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Review of arsenic contamination and human exposure through water and food in rural areas in Vietnam

Celia Hahn

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Review of arsenic contamination and human exposure through water and food in rural areas in Vietnam

Dissertation zur Erlangung des akademischen Grades

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Vorwort

Arsen ist ein Element, welches weit verbreitet ist und geogen in vielen Böden vorkommt. Es wirkt in vielerlei Hinsicht toxisch und daher wurde Ende der 80er Jahre in Deutschland der Grenzwert von 40 µg/L auf 10 µg/L im Trinkwasser gesenkt. In Europa ist das Grundwasser aus dem Trinkwasser gewonnen wird, wenig mit Arsen belastet, so dass Arsen hier nur eine untergeordnete Rolle spielt. In Südostasien hingegen sind die Arsengehalte in den Böden um ein vielfaches höher und damit die Gefahr, dass Arsen ins Trinkwasser gelangt wesentlich grösser.

Die Dissertation von Frau Hahn befasst sich mit dem Vorkommen von Arsen in Böden, Grundwasser und landwirtschaftlichen Produkten in Südostasien. Das Grundwasser und - bei ungenügender Aufbereitung - auch das daraus gewonnene Trinkwasser weist in diesem Teil der Erde häufig sehr hohe und weit über den Grenzwert von 10 µg/L Trinkwasser liegende Konzentrationen auf. Als Folge treten besonders in den ländlichen Gebieten durch Arsen im Trinkwasser und in den Lebensmitteln verursachte Krankheiten wie beispielsweise ‘blackfoot desease’ auf.

Frau Hahn beschreibt in ihrer Arbeit die Ursachen für das Vorkommen von Arsen im Grundwasser und den Weg dieses Schadstoffes in die Nahrungskette am Beispiel des Handwerksdorfes Dai Lam im Norden Vietnams. Dieses Dorf steht für tausende solcher Dörfer in ganz Südostasien und die Befunde können daher generalisiert werden.

Aus der Vielzahl der Untersuchungen wurde eine belastbare Datenbasis über das Arsenvorkommen in Wasser, Boden und Lebensmitteln in diesem Dorf erstellt und die Befunde mit den Ergebnissen von Untersuchungen zu diesem Thema in anderen arsenbelasteten Gegenden zu verglichen.

Sie konnte den Beweis erbringen, dass der Anstieg der Arsenbelastung im Grundwasser und damit in der Nahrungskette durch die Intensivierung der Landwirtschaft verursacht wird. Durch die Intensivierung wird bewirkt, dass das geogen an den Boden gebundene unlösliche 5wertige Arsen durch das Absenken des Redoxpotentials in das mobile und wesentlich toxischere 3wertige Arsen reduziert wird. Diese Tatsache ist besonders im Hinblick auf die Umstellung der Landwirtschaft von 2 Ernten Reis pro Jahr auf 3 Ernten pro Jahr kritisch.

Die Schlussfolgerungen aus dieser Arbeit stellen einen wesentlichen Baustein fuer die Entwicklung eines Decision Support System für die Entscheidungsträger in den vietnamesischen Behörden dar.

Ich wünsche Frau Dr. Celia Hahn auch weiterhin viel Erfolg in Ihrer wissenschaftlichen Karriere und freue mich, dass sie auch in den nächsten Jahren im Umweltschutzbereich in Vietnam tätig sein wird.

Prof. Dr. rer. Nat. Dr. h. c. Peter Werner

Abstract

The Red River Delta in Vietnam is one of the regions whose quaternary aquifers are polluted by arsenic. Chronic toxification by arsenic can cause severe illnesses such as cancer, skin lesions, developmental defects, cardiovascular and neurological diseases, and diabetes. In this study, a food processing craft village in the Red River Delta was investigated regarding the potential risk faced by the population due to arsenic. The potential sources of arsenic are the groundwater, the crops grown in the surroundings, and animal products from local husbandry. However, the occurrence of arsenic in nature is variable, and its bioavailability and toxicity depend very much on its specification: trivalent compounds are more toxic and often more mobile than pentavalent compounds, while inorganic species are generally more toxic than organic ones. Local conditions, such as the redox potential, strongly influence its specification and thus potential bioavailability.

The introduction to this work elucidates the key factors which potentially cause human exposure to arsenic: the geological setting of the study area, land and water use patterns, and the current state of research regarding the mobilization, bioavailability and plant uptake of arsenic.

Although the study area is located in a region where the groundwater is known to be moderately contaminated by arsenic, the level of arsenic in the groundwater in the village had not previously been determined. In this study, water use in the village was examined by a survey among the farmers and by water analyses, which are presented in the following chapters. Four main water sources (rain, river, tube well and a public municipal waterworks) are used for the different daily activities; the highest risk to human health was found to be the bore well water, which is pumped from the shallow Holocene aquifer. The water from the bore wells is commonly used for cleaning and washing as well as to feed the animals and for food processing. Products like noodles and rice wine were examined as well as local pork and poultry. Vegetables from the gardens and rice plants from the surrounding paddy fields were sampled and analyzed. All plants were found to have accumulated arsenic, leafy vegetables showing the highest arsenic concentrations.

The results are discussed and compared, and conclusions are drawn in the last part. The reducing conditions in the paddy fields are likely to have a strong influence on arsenic uptake in rice plants and on transport to the aquifer. The installation of a wastewater treatment plant under the research project INHAND, which was funded by the BMBF German Ministry of Education and Research, led to lower arsenic concentrations in the groundwater.

Soaring industrialization, the growing population, and the consumers’ changing behavior will widely affect land and water use and hence the potential mobilization of arsenic. In order to mitigate further human exposure to arsenic, wastewater needs to be treated and the reducing conditions in the rice fields need to be decreased by means of enhanced cultivation methods.

Zusammenfassung

Das Delta des Rotens Flusses in Vietnam gehört zu einer der Regionen deren quartäre Grundwasserleiter durch Arsen belastet sind. Eine chronische Arsenvergiftung kann schwere Krankheiten wie Krebs, Hautwunden, Entwicklungsdefizite, Herzkreislauf- und neurologische Krankheiten und Diabetes hervorrufen. In dieser Arbeit wurde ein Nahrungsmittel produzierendes Handwerksdorf im Delta des Roten Flusses hinsichtlich des Gefährdungspotentials der Bevölkerung durch Arsen untersucht. Die möglichen Arsenquellen sind das Grundwasser, die Gemüse und Getreideernten der umliegenden Felder und die Produkte der örtlichen Tierzucht. Arsen kommt in der Natur in sehr unterschiedlichen Formen vor und die Bioverfügbarkeit, sowie die toxische Wirkung hängt stark von der Spezifizierung ab: dreiwertige Verbindungen sind meistens giftiger und mobiler als die fünfwertigen und anorganische Arsenverbindungen sind generell toxischer als organische. Die lokalen Bedingungen, wie z.B. das Redoxpotential haben einen beträchtlichen Einfluss auf die Spezifikation und damit auch auf die Bioverfügbarkeit des Arsens.

Der einleitende Teil der Arbeit beleuchtet die verschiedenen Einflussgrößen, welche potentiell zu einer Arsenaufnahme durch den Menschen führen können: die geologischen Gegebenheiten des Untersuchungsgebietes und Land- und Wassernutzungsstrukturen, sowie den Stand der Forschung auf dem Gebiet der Mobilisierung, Bioverfügbarkeit und Pflanzenaufnahme des Arsens.

Das Untersuchungsgebiet liegt in einer Region, deren Grundwasser mäßig durch Arsen verunreinigt ist, bislang gab es in dem Dorf jedoch keine Untersuchungen des Grundwassers hinsichtlich des Arsengehaltes. In dieser Arbeit wurde die Wassernutzung des Dorfes durch eine Umfrage und durch Wasseranalysen untersucht, welche in den folgenden Kapiteln dargestellt werden. Im täglichen Leben werden vier Hauptwasserquellen genutzt (Regen- und Flusswasser, Hausbrunnen und ein kommunales Wasserwerk). Es wurde festgestellt, dass die höchste Gefährdung durch die Nutzung der Hausbrunnen ausgeht, welche Wasser des flachen Grundwasserleiters fördert. Das Wasser der Hausbrunnen wird normalerweise zum Wachen und Reinigen genutzt, aber auch zur Versorgung des Nutzviehs und in der Produktion von Nahrungsmitteln in den Handwerkbetrieben). Die Produkte der der Betriebe, Reiswein und Nudeln, wurden untersucht, ebenso wie die Produkte der Viehzucht. Zudem wurde Gemüse und Reis der umliegenden Felder beprobt und analysiert, wobei das Blattgemüse die höchsten Arsenkonzentrationen enthielt.

Im letzten Teil der Arbeit werden die Ergebnisse der Untersuchungen diskutiert und zueinander in Bezug gesetzt. Die reduzierenden Bedingungen in den Reisfeldern haben einen sehr großen Einfluss auf die Aufnahme des Arsens in die Pflanzen aber vor allem auch auf die Transportprozesse in zum Grundwasser. Der Bau einer Pilotanlage durch das vom BMBF geförderte Forschungsprojekt INHAND führte zu geringeren Arsenkonzentrationen im Grundwasser.

Die schnell zunehmende Industrialisierung, der Bevölkerungszuwachs und das sich ändernde Konsumverhalten in der Region wird die Strukturen der Land- und Wassernutzung in den nächsten 30 Jahren stark verändern und damit zu einer weiteren Mobilisierung des Arsens beitragen. Um den Eintrag von Arsen in die menschliche Nahrungskette zu verhindern, muss eine flächendeckende Abwasserbehandlung und die Nutzung von fortschrittlichen Reisanbaumethoden vorangetrieben werden.

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Regarding the non-scientific side of my thesis, I would like to thank my son and my partner for their unremitting belief in my ability and their patience. Moreover, I would like to thank my whole family, my parents and my brother. And finally, I'd like to thank my friends who shared the ups and downs on my way from Madrid, Kassel, Brussels, Cologne, Bonn, Bielefeld, Dresden, Hanoi and Berlin. This work would not have been possible without their friendship, help and understanding.

Contents

List of abbreviations

ADHD	attention deficit hyperactive disorder
As	arsenic
As_{in}	inorganic arsenic
As_{tot}	total arsenic
b.w.	body weight
CONTAM	Panel on Contaminants in the Food Chain
CAC	Codex Alimentarius Commission
CAS	Chemical Abstracts Service
CTIC	Center for Training and International Cooperation of the Vietnam Academy for Water Resources
DO	dissolved oxygen
DMA	dimethylarsinic acid
DSMA	disodium methyl arsenate (DSMA)
dw	dry weight
EC	electric conductivity
FAO	Food and Agriculture Organization of the United Nations
GAS	golden apple snail
HFO	hydrous ferric oxides
INHAND	Integriertes Wasserwirtschaftskonzept für Handwerksdörfer am Beispiel von Dai Lam in Vietnam
MMA	monomethylarsonic acid
MSL	mean sea level
MSMA	monosodium methyl arsenate
NOAEL	non-observed adverse effect level
ORP	Oxidation-Reduction Potential
PTWI	provisional tolerable weekly intake
RRD	Red River Delta
SEF	sequential extraction frationation
SBR	sequencing bed reactor
TETRA	tetramethylarsonium ion
TMAO	tetramethylarsonium

UNICEF	United Nations Children's Fund
US EPA	United States Environmental Protection Agency
ww	wet weight
WHO	World Health Organization

List of figures

List of tables

1 Scope of this work

In the Red River Delta in the north of Vietnam, about 11 million people are potentially affected by increased arsenic concentrations in the quaternary groundwater. The center of contamination is situated southeast of Hanoi, where the concentrations in the groundwater exceed the WHO standard of 10 µg/L up to 80-fold. Little attention has been paid to the peripheral zone of the arsenic contamination. In this study, a traditional craft village in the northern Red River Delta was investigated regarding the villagers' exposure to arsenic, manganese and iron through nutrient uptake. In previous studies, this area had been found to be moderately affected by arsenic in the groundwater (Winkel et al. 2011). However, temporal and small-scale spatial variations in the groundwater gave rise to the assumption that the question of arsenic exposure has to be considered as a variable, complex problem. In addition to uptake through water, the locally grown food and animal livestock are potential sources for As and heavy metals. However, since only little information is available about the level of As in food in Vietnam, several crops, types of meat and other food were sampled and analyzed.

An additional task of this study is the question of how much the daily rural activities can affect the mobilization of arsenic and what the possible pathways and mechanisms could be. In this regard, a review was carried out taking into account scientific studies from other affected areas in India, Bangladesh and China.

Vietnam's economy is in transition and industrialization is proceeding fast. The socio-economic changes imply strong interventions in the land and water use structure of the country. The changes have already been set in motion by national target programs and will lead to a loss of arable land and hence to intensified farming. This study considered the possible impact of these developments on the availability of arsenic.

2 Introduction

The current environmental situation in the Red River Delta has various aspects and influences which need to be considered from an interdisciplinary point of view. The geographical and geological setting provides the natural framework for the environmental situation. The fact that large parts of the area bear high arsenic concentrations in the sediments and aquifers is the cause of serious concern. The arsenic-containing sediments originate from the Himalayan massif, where the parent rock material was degraded by dilution processes and erosion. Human activities such as wastewater drainage in the receiving streams, changes of the groundwater table and the use of fertilizer led to changing physicochemical soil properties and to the possible mobilization of arsenic. Given this background, the social, demographic, industrial and rural development contains both opportunities and threats for human health and the environment.

This section examines the current situation in regard to the natural framework, the origin of the arsenic, the status of the rivers, and its influence on the aquifer. Additionally, the uptake processes of As in plants as studied by many international scientific working groups are explained. The special conditions of rice cultivation in paddy fields doubtless have an important influence on the As concentration in rice plants and rice grains. Furthermore, the legal framework is described along with the upcoming measures of the Vietnamese government to protect the natural resources and human health.

2.1 Geographical and geological setting of the study area

The Red River Delta is part of a huge river system in northern Vietnam with a catchment area of 169,000 km², which stretches as far as China and Laos. The river system includes the Red River and its tributaries, which is why the name ‘Red-Thai Binh River system’ is widely used (figure 1). The plain of the delta is surrounded by mountain massifs, where the sediments in the delta originated. These massifs include the Hengduan massif in China, a branch of the Himalaya massif, the Hoang Lien Son massif in Vietnam, and the mountains of Nam Neun in Laos. The total area of the river delta is 22,000 km², making it four times as big as the Danube Delta in Europe.

The Delta is a plain with many, partly very small and sharp elevations, which form landscape scenery of parts of the Delta like the picturesque Halong Bay.

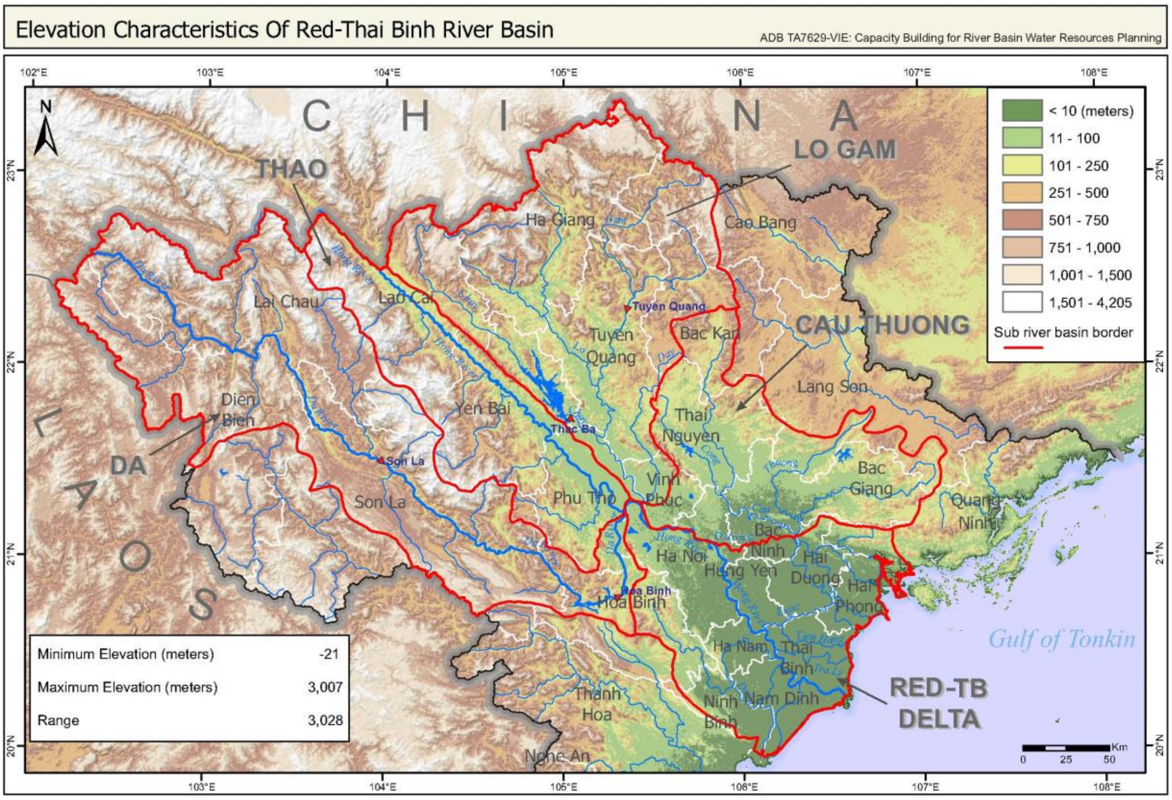

Figure 1: The Hong/Thai Binh River system (ADB 2012)

The geological structure of the Delta is characterized by intensive tectonic activity, which has been related to the plate tectonic movements since the Miocene. The northward drifting Indian plate induces tectonic stress in the northern East Asian region around Tibet. During the thrust folding, a set of fault zones developed. One of the most prominent zones is the Red River fault zone (Figure 2) (Schärer 1990, Wysocka 2003). This zone runs NW–SE from China into the region of the Red River Delta. The present area is subject to sequenced subsidence of currently 0.04 to 0.12 mm/a (Tanabe 2006, Tanabe 2003), which leads to the varying sediment layers of the Neogene and Pleistocene. The uplifting area of the Himalaya massif has provided huge quantities of sediment material.

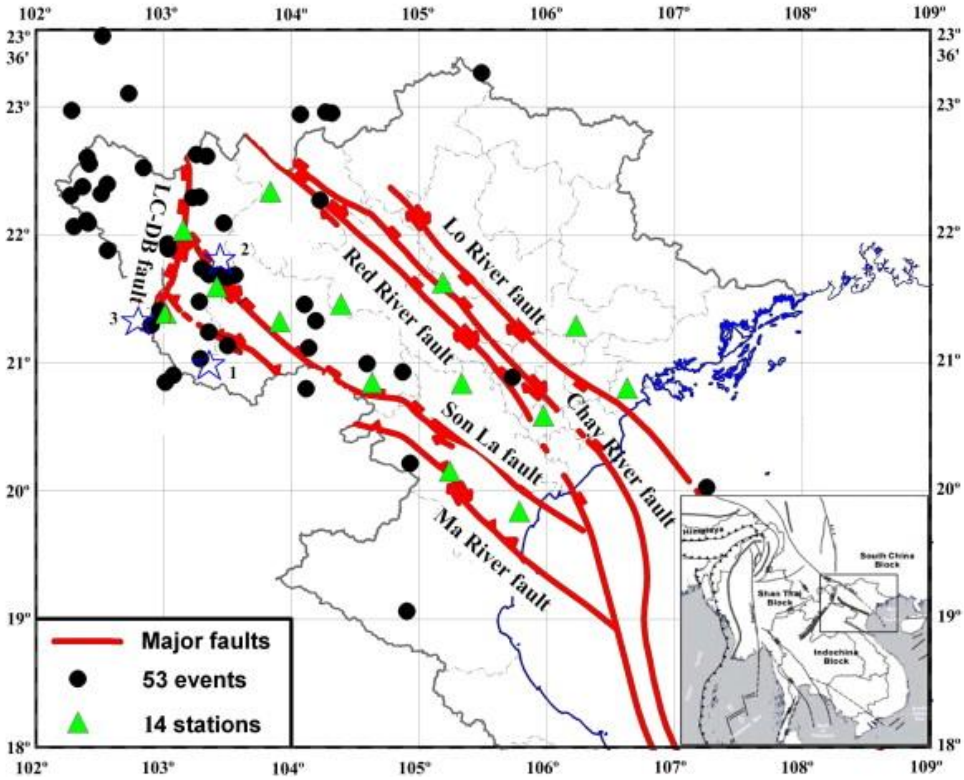

Figure 2: Map of the major fault systems in the Red River Delta (Nguyen et al. 2012)

The present hilly and mountainous boundary of the delta consists of crystalline bedrock from the Proterozoic and from Precambrian and Mesozoic rocks.

The hills mentioned in the area consist of coarse sediments such as conglomerates of white-grey quartz and silty-calcareous layers of the Hòn Gai formation; these are part of the transgression sediments (Nor-Räth), which can have a thickness of 900 m.

The Mesozoic layers are discordantly superposed by marine Pleistocene layers. These fluvial and alluvial yellow-greyish clay and silt layers are assigned to the Lechi-Hanoi, Hanoi and Vinh Phuc formation, which are widely present in the study area and have a thickness of up to 100 m in the Hanoi area. The layers were probably formed under colder climate conditions when the sea level was lower than today (Mathers 1999).

The lower and middle Holocene sediments of the Hai Hung Formation are likewise widespread; they consist of dark clay and silt layers typical of lacustrine marshy sediments.

The upper Holocene layers of the Thai Binh formation have two facies in the study area: near bigger rivers and lakes they consist of sandy mud and in the other areas these layers consist of silty clay, which is often used for brick manufacturing (Mathers 1999, Hoang Ngoc Ky 2001).

The area is characterized by cultural landscapes. Rice cultivation led to the development of a complex irrigation and drainage system. Rivers, ponds and canals are interconnected; the area is structured by dams and paddy fields (Bakker et al. 2003).

2.2 Hydrological situation

The hydrology of the Red River Delta is a complex system of several rivers and their tributaries as well as canals, dams and the tidal influence of the eastern Vietnamese sea. The fluvial-lacustrine sediments bear several aquifers of varying quality.

2.2.1 Surface water

The eponymous Red River has a length of 1,126 km, 216 km of which flows through the Delta (from Son Tay to the coast). In the Delta, the Red River is split into two branches: Day River and Duong River. The whole system consists of two main branches, the Red River Branch consists of Duong River, the Thai Binh River Luoc, Chau Giang, Dao, Nhinh Co and Dao River (Tran, 2007Dang, Fontenelle 1995, Fontenelle 1997, Ritzema 2008).

The Day River, the other part of the system forming the Delta, is a shallow river which is threatened by siltation. The five main tributaries are the Bui, Nhue, Chau, Boi and Dao rivers.

The characteristic paddy fields, canals, ditches and dams are essential for the socio-economic system in northern Vietnam. All in all there are 4,500 km of dams, of which 3,000 km are river dams and 1,500 km protect the coast line (World Bank et al. 2003).

The central part of the Delta is a plain with a mean height of 2–17 m MSL.

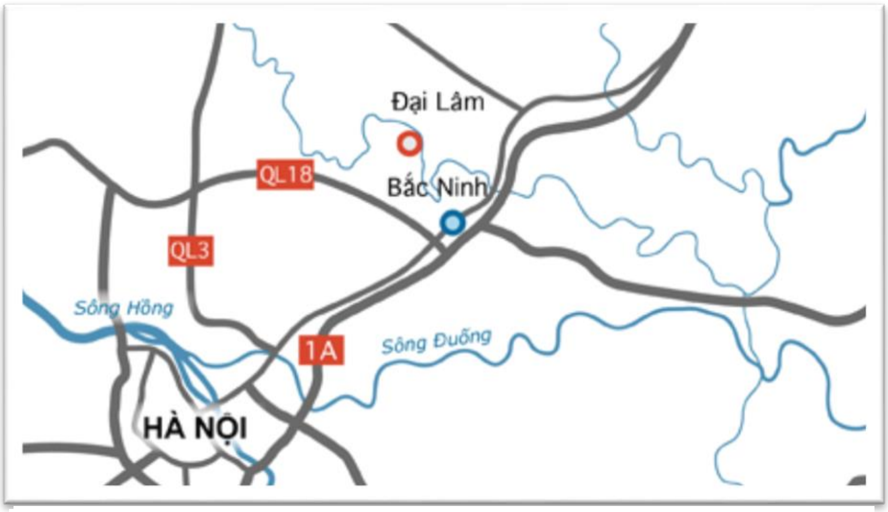

Figure 3: Map of the area

Dai Lam, the study area (Figure 3) is situated on the rim of the Cau River, one of the most polluted rivers in Vietnam (Figure 4). The Cau River is the main river of the Thai Binh River system. It originates from Phia Deng Mountain at an altitude of 1,527 m in the southeast Piabioc Mountains. The length of the Cau River is 288 km. The river network in the Cau River basin is relatively complex with evenly distributed tributaries. The catchment area is 6,030 km². The annual rainfall in the Cau River basin varies from 1,400 to 2,700 mm (average: 1,680 mm). The rainy season in the Cau River basin usually occurs from May to September in the upper part of the basin, extending to October in the middle and lower parts. Rainfall in the rainy season makes up about 65–85% of the annual rainfall.

2.2.2 Impact of human activities on surface water quality and distribution

The area of the Red River Delta is one of the oldest cultural landscapes in the world. For thousands of years, the land has been shaped by human activities such as agriculture, horticulture und aquaculture, resulting in the construction of thousands of dams, canals and ditches (Fontenelle 1997). Intensive agriculture craft processing and trade have led to one of the highest population densities in the world (Fontenelle 1997).

Despite being one of the granaries of Vietnam, the Cau River region is increasingly being transformed into an industrialized area. More than 4.5 million people live in the area and the population density is 922 persons/km^{2,} which is twice as high as the average in Vietnam (427 person/km^2). Although incomes have risen due to increasing industrialization over the last decade, very little has been invested into the infrastructure to protect the environment and natural resources. This is in spite of the fact that economic development is putting high pressure on natural resources and will probably continue to do so during the next few decades.

Like many other rivers which flow through industrialized areas, the Cau River is affected by several factors.

Sand mining:

Due to increasing industrialization, sand mining activities in the Cau River have boomed since the turn of the millennium. Since 2000, the daily extraction volume has quadrupled. Many illegal mining activities have been reported. As a result, the dams and river banks are subject to erosion and the water tables have been lowered (Monre 2007).

Mining activities in the upper river stream:

The Cau River passes through four provinces as well as several cities and industrialized areas. The upper Cau River and its tributaries are strongly affected by mining activities; the lower Cau River receives high concentrations of organic pollutants, suspended solids and oil waste from small scale industries and domestic activities. The most polluted section is probably downstream of Thai Nguyen City due to untreated wastewater released by heavy industrial factories, paper mill, mining (gold and coal mining) and agricultural activities along the river (Monre 2007, Molle, Hoanh 2008).

Craft villages from all branches are situated on the rim of the stream. Domestic wastewater is not treated, which probably poses one of the major problems both now and for the future.

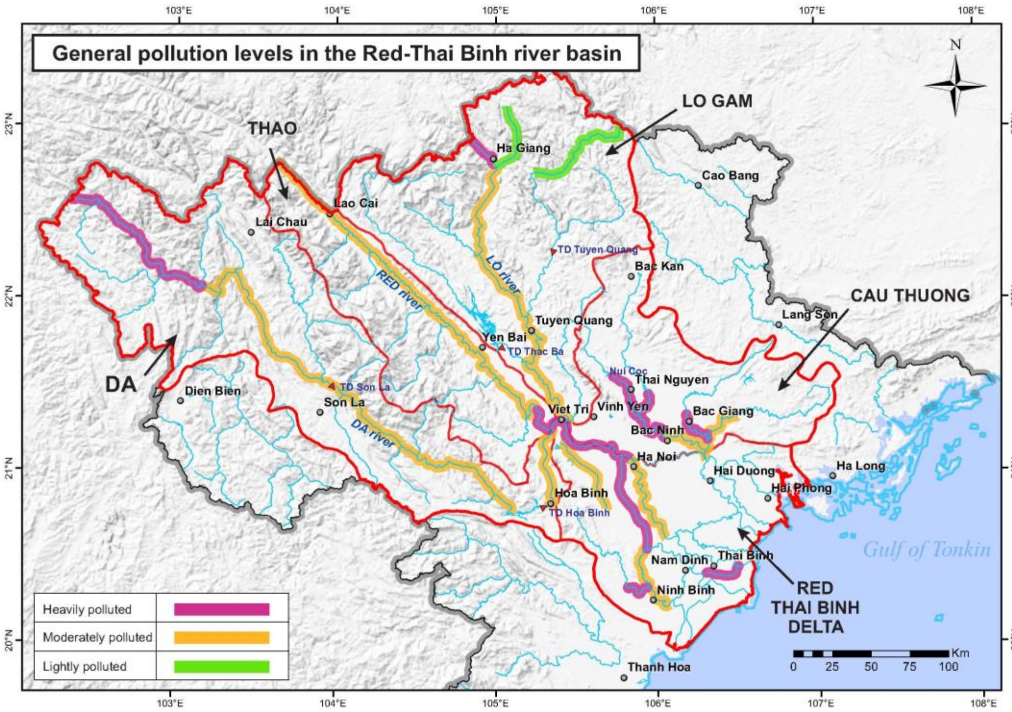

Figure 4: Pollution levels in the river system (ADB 2012)

2.2.3 Hydrogeology

In the Red River Delta, groundwater is the main source for domestic water use, and throughout the Delta, several aquifers are exploited (Bui 2012). The main aquifers for water procurement are embedded in the mid-upper Pleistocene layers of the Vinh Phuc–Hanoi formation, which consist of clay sands, sands, gravels and cobbles. The overlying Hanoi formation consists of coarse-grained quartz gravel and sand, the underlying layers are fine-grained (Vinh Phuc formation) sandwiching the Holocene-Pleistocene aquitard (Bui 2011). The thickness of the aquifer system of the delta can be up to 100 m in coastal regions, and increases from the northwest to the southeast of the delta (Bui 2012).

2.3 Arsenic occurrence

Arsenic is a metalloid listed in the periodic system of the elements in the fifth main group. Its atomic number is 33. Elemental arsenic only occurs in very small quantities in ores, yet is present almost everywhere, mostly as sulfur or oxygen compounds. In the lithosphere, it is the fifty-third most abundant element. However, As is also present in the hydrosphere, atmosphere and biosphere. Although As is found as a trace element in all living organisms, it isn't an essential element. The occurring oxidation states are –3, 0, +3 and +5, although in nature its predominant oxidation states are +3 and +5.

As has a wide range of organic and inorganic species. To date at least 200 organic As compounds have been detected in natural environments, and 565 As-containing natural minerals are known (Barthelmy 2011) with different As concentrations.

The most prominent organic compound is the highly toxic cacodylic acid, also known as Agent Blue, which was used as herbicide in the Vietnam War (Stellman et al. 2003). Arsenosugars or arsenoribofuranosides (ribose derivatives) and arsenobetains are very common in algae and crustaceans (Francesconi, Edmonds 2001, Niegel, Matysik 2010). The inorganic species of high environmental and human health relevance are the trivalent aresenite and pentavalent arsenates.

The toxicity and the environmental fate of As depend on the species and vary from harmless (arseosugars) to highly toxic like arsenite, which has been used as raticide, and cacodylic acid, which was used as a biological weapon in the Vietnam War.

In the historical context, As came to prominence as a pharmaceutical as long ago as at the end of the Zhou Dynasty (222 BC) (LIN 1978, Matschullat 2000), while Albertus Magnus synthesized As in 1250 AD (Schröter W. et al. 1983, Marsh 1837). In Europe, As was described as the ‘king of poisons’ because only small quantities of the odorless white power (arsenite) were sufficient to be used as an effective homicidal and suicidal agent. In 1832, James Marsh found a reliable method to detect As (Marsh 1837), and from then on it was easier to prove crimes related to the ingestion of As.

Robert Bunsen (1811–1899) studied the composition of cacodylic acid (from the Greek kakodhs – ‘stinking’) and found “the smell of this body produces instantaneous tingling of the hands and feet, and even giddiness and insensibility.... It is remarkable that when one is exposed to the smell of these compounds the tongue becomes covered with a black coating, even when no further evil effects are noticeable.” (Bunsen 1843) Losing the sight of his right eye in a laboratory accident, he was unable to complete his studies. However, one of the main outcomes was that the toxicity of As depends on the species, which was an important step towards understanding the element’s toxicology (Debus 1903).

2.3.1 Arsenic toxicity

Arsenic exposure in humans can occur through different pathways such as inhalation, dermal absorption, and the ingestion of food, water and soil. Acute arsenic poisoning leads to nausea, vomiting, abdominal pain and severe diarrhea. Encephalopathy and peripheral neuropathy have been reported (Ratnaike 2003). Chronic arsenic toxicity evokes multisystem disease and is a human carcinogen affecting numerous organs, a condition known as arsenicosis (Ratnaike 2003). Its toxicological effect depends on the specification of As (Table 1). Trivalent compounds are generally more toxic than pentavalent compounds, organic compounds are less toxic than inorganic ones, and the toxicity decreases with increasing methylation (with the exception of the tetramethylarsonium ion).

Organic As species

Arsenobetain and arsenocholine are regarded as harmless to human beings (ATSDR (Agency for Toxic Substances and Disease Registry) 2007, Borak, Hosgood 2007). Currently, the effects of arsenosugars and arsenolipids are being studied because arsenosugars may be metabolized to more harmful arsenic compounds (Andrewes et al. 2004).

Trimethylarsinoxid (TMAO) and tetramethylarsonium (TETRA) are considered moderately toxic (Contreras-Acuna et al. 2013). Methylated species like MMA^{+V} and DMA^{+V} are also considered moderately toxic (Fattorini, Regoli 2004). However, in mammals, pentavalent methylated As compounds are metabolized to trivalent compounds, which are regarded as highly reactive and responsible for intoxination (Watanabe et al. 2002). Recently, another As compound has attracted interest because of its proved toxic properties in bladder cells: thiodimethylarsinic acid (Ebert et al. 2014).

Table 1: As species, CAS numbers and reported toxicity

Name	Formula	CAS number	Oxidation state	EU classification	LD_{50} (mg/kg)
Arsine	AsH_3	7784-42-1	+III	(F+) (T+) (Xn) (N)	0.003 (mouse)[1] Fatal dose: 250 mg/m^3 30 min[2]
Arsenous acid	H_3AsO_3	13464-58-9	+III		0.031 (rat)[3]
Arsenic acid	H_3AsO_4	7778-39-4	+V	(T) (N)	0.048 (rat)[3]
Arsenic trioxide	As_2O_3	1327-53-3	+III	(T+) Carc. Cat. 1 (N)	0.014 (rat) 0.0014 (human)
Arsenic pentoxide	As_2O_5	1303-28-2	+V	(T+) Carc. Cat. 1 (N)	0.008 (rat)[4]
Arsenic trisulfide	As_2S_3	1303-33-9	+III	(T) (N)	0.254 (mouse)[3]
Sodium arsenite	$NaAsO_2$	7784-46-5	+III	(T) (N)	0.041 (rat)[5]
Sodium arsenate	Na_2HAsO_4	7778-43-0	+V	(T)	0.001-0.02 (mouse)[5]
Cacodylic acid	$C_2H_7AsO_2$	75-60-5	+V	(T) (N)	0.644[6]

[1] LEVVY, G. A. *The toxicity of arsine administered by intraperitoneal injection*. pp. 287–290
[2] Mayer, D. R.: Essential trace elements in humans. Serum arsenic concentrations in hemodialysis patients in comparison to healthy controls. Biol. Trace Elem. Res. 37, 27 (1993).
[3] Material safety data sheet http://www.t3db.ca/
[4] Material Safety Data Sheet http://www.sciencelab.com/
[5] wikipedia
[6] Fischer scientific data sheet: http://www.fishersci.com/

Table 1 continued

Name	Formula	CAS number	Oxida-tion state	EU clas-sification	LD_{50} (mg/kg)
Monomethylarsonic acid	$CH_3H_3AsO_3$	124-58-3	+V		1.8 [7]
Monomethylarsonous acid	CH_5AsO_3	25400-23-1	+III		
Dimethylarsinic acid	$C_2H_7AsO_2$		+III		1.2 [7]
Arsenobetaine	$C_5H_{11}AsO_2$	64436-13-1		(T) (N)	>10 [7]
Arsenocholin (AC)	$C_5H_{14}AsO^+$	39895-81-3		(T) (N)	>6.5[7]
Arsenosugars	Group				
Trimethylarsinoxid (TMAO)	C3H9AsO	4964-14-1	+III	(T) (N)	10.6[7]
Tetramethylarsonium (TETRA)	$C_4H_{12}As$		+V	(T) (N)	

2.3.2 Risk potential of arsenic in diet

No international compulsory threshold for arsenic in food has been defined yet. In 1981, the WHO postulated that a daily intake of arsenic of more than 1 mg/kg would lead to skin irritations, but it was soon realized that this value was too high. In 1993, the Codex Alimentarius Commission (CAC), a joint scientific board of FAO and WHO, defined a value for the PTWI (provisional tolerable weekly intake) of 15 µg/kg body weight. For an adult weighing 75 kg, this would be a daily maximum value of 112,5 µg. This value has been recognized as too high in general. In 2009, the CONTAM panel of the EU recommended defining a lower value for the PTWE because values between 0.3 and 8 µg/kg b.w. per day were thought to increase the risk of cancers of the lung, skin and bladder as well as skin lesions (EFSA 2014, EFSA CONTAM Panel 2009).

The CAC is currently drawing up a new standard for arsenic in rice. The difficulties result from the different arsenic species which have different toxicological effects and from the different analytical methods. Although several approved methods for analyzing arsenic in food exist across the world, most of them only deliver the total arsenic concentration.

The CAC report from 2012 names two thresholds:

0.2 mg/kg As_{in} and 0.3 mg/kg As_{tot}

In both Asia and Europe, the bulk of consumed arsenic results from rice intake. However, mean daily rice consumption varies enormously from 9 g in Europe to 278 g in Asia.

The EFSA report from 2014 used 114,200 results (gathered between 2003 and 2013). In Europe, exposure to inorganic As (As_{in}) in the adult population ranged from 0.13 to

[7] SHIOMI, Kazuo. *Arsenic in marine organisms: Chemical forms and toxicological aspects.* pp. 261; 261–282; 282

0.56 µg/kg b.w. per day for average consumers, and from 0.37 to 1.22 µg/kg b.w. per day for 95th percentile (high-level) consumers. The dietary exposure of children under three years was generally estimated to be 2 or 3 times higher than that of adults (EFSA 2014). In Europe, no general threshold for As in aliments has been defined yet. The standard of 1.0 mg/kg in Spain and United Kingdom dates back to the 1970s and the 1950s, respectively.

Australia and New Zealand

The Australia New Zealand Food Standards Code defines a set of maximum values for As in aliments:

Grains: total As 1.0 mg/kg

Crustaceans and fish: As_{in} 2.0 mg/kg

Mollusks and edible algae: As_{in} 1.0 mg/kg

China

The Chinese Food Standards Agency (2005) has defined Maximum Contaminant Levels (MCLs) for inorganic As in rice grains at 0.15 mg/kg, for edible algae at 1.5 mg/kg, and for aquatic animal products at 0.5 mg/kg (FAO/WHO 2011).

2.4 Arsenic contamination in the groundwater resources of the Red River Delta

"The contamination of groundwater by arsenic in Bangladesh is the largest poisoning of a population in history, with millions of people exposed." These were words that Allan H. Smith chose in 2000 to introduce the results of his study of Bangladesh (Smith et al. 2000). Since the 1940s, thousands of wells have been sunk in the Ganges-Brahmaputra Delta in order to ensure a safe drinking water supply for the population. The delta is one of the biggest deltaic regions in the world and, like many delta regions, is densely populated. Since the 1980s, well construction has been funded by the WHO. First evidence of skin irritations due to As uptake were reported in 1983 (Saha 1984, Smith et al. 2000). In the early 1990s, groundwater analyses were carried out which confirmed the high arsenic concentration in many wells. National (Ahmad et al. 1997) and international (British Geological Survey 2001) studies showed that in 30% of the wells, the water exceeded the former international standard of 50 µg/l As.

In 2008, UNICEF published a report on the arsenic issue after 4,750,000 wells had been tested. A total of 1,400,000 wells were found to be contaminated and the number of people exposed to contaminated drinking water daily was estimated to be 20 million (UNICEF 2008).

A similar program for sinking wells in the rural areas of Vietnam was conducted in the 1980s. The aim was to install a safe drinking water supply for the rural population. In 2001, Michael Berg published the results of a detailed study carried out in Hanoi and surroundings. The majority of the wells revealed high arsenic concentrations. The mean arsenic concentration was 159 µg/l and the highest value was 3,050 µg/l (Berg et al. 2001a). In the following years, several point analyses were carried out (Agusa 2002, Agusa et al. 2006, Jessen 2008, Eiche 2008, Brammer et al. 2009, Eiche 2009). In 2011, the results of a structured investigation were published (Winkel et al. 2011): 510 household wells were sampled and analyzed. Based on these data, a risk map was modeled. It is assumed that about 3 million people in the Red River Delta – about 27% of the population – are exposed to contaminated groundwater. In contrast to the Ganges-Brahmaputra Delta, only a few health studies have been conducted and published for Vietnam. (Berg et al. 2007a, Agusa et al. 2010, Agusa et al. 2014, Agusa et al. 2006) analyzed hair samples of people living in affected areas (As concentration in groundwater > 50µg/l) in the Red River Delta. The results ranged from 0.2 to 2.75 mg/kg (Berg et al. 2007b), 0.09 to 2.8 mg/kg (Agusa et al. 2006) and 0.07–7.51 mg/kg (Agusa et al. 2014), which is higher than 1 mg/kg, indicating the level of arsenic intoxication diseases. This reveals that major parts of the population in the affected areas have an enhanced arsenic uptake risk. Very few cases of arsenic-related diseases have been reported from the Red River Plain. The reason proposed by Berg et al (2007) is that arsenicosis is difficult to diagnose, especially in the early stage. Furthermore, the general nutrition of the population in Vietnam might be better than in other affected regions like Bangladesh. Agusa et al. (2014) raised the idea of an ethnological resistance towards arsenicosis.

Regions in Cambodia (Berg et al. 2007, Polya 2004, Feldman et al. 2007), Thailand (Williams 1996), Myanmar (Winkel 2008), Sumatra (Winkel 2008a), China (Guo et al. 2003), Mongolia (Guo et al. 2001)and Laos (Chanpiwat et al. 2011) are also affected by high arsenic concentrations in groundwater.

2.4.1 Occurrence and origin of arsenic in the Red River Delta

High arsenic values in groundwater samples have been reported from the lower Holocene aquifer as well as from the lower Pleistocene aquifer (Berg et al. 2001b, Buschmann et al. 2007, Berg et al. 2007, Berg 2008, Giger 2003, Winkel 2008, Jessen 2008, Postma 2007). Though anthropogenic contamination of the groundwater due to the use of pesticides and mining activities could be the source of the arsenic contamination, it is evident that the extensive groundwater contamination by arsenic is geogenic: the quaternary sediments contain considerable arsenic concentrations in the moment of the sedimentation in the Red River plain. There are indications that the sediments are derived from the Himalayan massif in the northwest of the Red River Plain, but since no detailed investigation has been carried out into the origin of the sediment, no information is available about the original minerals or the rocks' composition (Jessen 2008, Postma 2007). Based on studies carried out in West Bengal and Bangladesh, it can be assumed that the arsenic was originally bonded to minerals like arseopyrite or serpentinite (Li 2013a). Mechanical erosion, dissolution and oxidation processes of arsenic-bearing rocks led to the transport of arsenic to the plain, where high concentrations are bonded to iron(hydr)oxides in the form of pentavalent arsenate (Figure 5).

Due to its bonding affinity, As(V) is less mobile than As(III), which is more toxic. Therefore, under specific conditions, the arsenic concentration in the sediment doesn't pose a severe problem to public health. However, by changing the redox conditions within the sediments and aquifers, arsenic may be reduced to As(III).

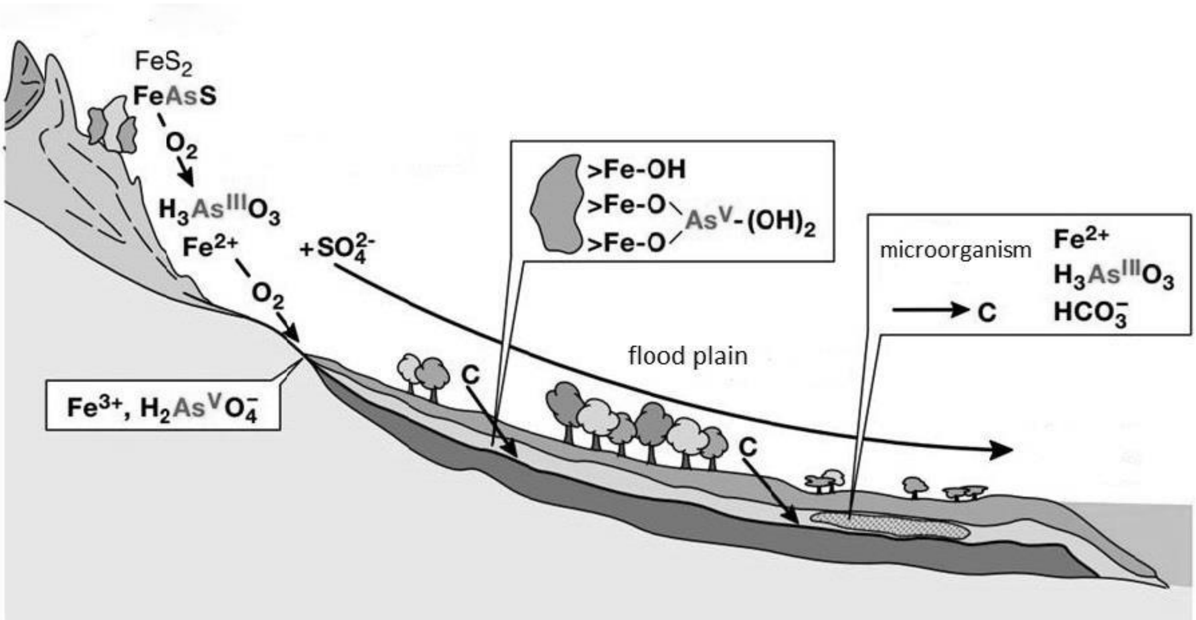

Figure 5: Development of the Red River flood plain sediments and arsenic dissolution and redeposition in the sediments (after Hug 2001)

2.4.2 Mobilization processes

To obtain a better understanding of the processes responsible for the mobilization of arsenic, many studies have been conducted on the bonding and releasing conditions. It is generally assumed that most of the arsenic in soils is adsorbed or bonded on iron oxides (magnetite or haematite), iron hydroxides (limonite, goethite) or amorphous iron oxides (Dixit, Hering 2003). Although other minerals tend to absorb arsenic, such as micas, clay, sulfides and carbonates (Chakraborti et al. 2001, Goldberg 2002a, Polizzotto 2006), the sorption processes of the iron(hydr)oxides and their effects on arsenic mobilization are probably the most studied processes and several theories are discussed in the scientific literature. The mechanisms and preconditions for the release of arsenic in the aquifer are understood in many respects, but the problem is complex. Since the turn of the millennium, several investigations have been carried out in order to develop a deeper understanding of the releasing processes:

- The oxidation of arsenical pyrites is the key process in the release of As from the primary minerals and thus for the presence of As in river basins such as the Ganges and the Red River Basin. Additionally, oxidation of sediment-bonded As can occur as a result of excessive water pumping and lowering the water table (Chakraborti et al. 2001, Sikdar 2001) and can induce the mixing of oxidizing and reducing aquifer water (Tareq et al. 2003). However, in most As-bearing regions, SO_4^{2-} concentrations were too low to attribute As mobilization to oxidizing processes.

- The most widely accepted theory is the microbially induced reduction of ferric(hydr)oxides releasing arsenic from aquifer sediments (Nickson et al. 1998, Harvey et al. 2002, McArthur 2001a, Dowling 2002). Oxidizing conditions and the presence of Fe induce inorganic arsenic to bond to FeOOH coatings on soil particles. Reducing conditions resulting from organic carbon oxidation can lead to the microbial dissolution of FeOOH coatings. This leads to the release of Fe^{2+}, As(III) and As(V).
- Wang and Mulligan (2006) proved the influence of naturally occurring organic matter on sorption behavior by interacting with mineral surfaces and/or with As itself. Naturally occurring matter thus may play a substantial role in the release of As from soil into groundwater. Similar findings were also published by Bauer, Blodau (2006a).
- Arsenic is often adsorbed on iron-containing minerals like goethite and haematite and competes with other anions like phosphates, sulfates and molybdates (Smith et al. 2002).
- The carbon source for continuing microbiological activity could be derived from peat deposits in the Holocene sediments (McArthur 2001b) or from surface water. Neidhardt (2014) found that a rise in organic carbon (by inducing succerose) in an aquifer increases the microbial activity and the concentration of dissolved Fe, but the study also postulated a considerable buffer capacity for bonded arsenic because the arsenic increase in the groundwater was much lower than expected.
- The solid phase arsenic concentration in the aquifer sediments is likely too low to be the source layer of the arsenic (Smedley 2002a). Therefore, the mobilization and transport of arsenic from the uppermost layers is postulated (Ahmed 2004, Thi Hoa Mai 2014).
- Appelo et al. (2002) and Anawar (2003) postulated the release of arsenic species by HCO_3^-, which might be formed from carbonates in the sediments.
- In a study site in Bangladesh, (Polizzotto 2006) found that 60% of the arsenic was bonded to sulfides and therefore reducing conditions do not affect the mobility of arsenic in this case. Instead, Polizzotto suggested a cycling process from oxide-bonded to sulfide-bonded arsenic. This cycle depends on seasonal variation and irrigation activities: in the wet season, reducing conditions predominate, provoking the release of arsenate from the sediment particles and adsorption on sulfides; in the dry season, the conditions become oxidizing: arsenic may be released and adsorbed on iron oxides.
- Weng et al. (2009) found out that arsenic desorption from iron oxides has a correlation with humic and fluvic acids because of the competition between humic and fulvic acids with arsenate.

- Phosphate and arsenate have a similar structure as well as similar dissociation constants for their acids and solubility products for their salts. Hence both components compete for the sorption components. Adriano (2001) detected the increased mobility of arsenic in soil after the application of P-containing fertilizers.
- The application of dissolved organic carbon into the soil may also play a role in the mobilization of arsenic because of the increased microbial activities (Harvey et al. 2002).
- Polizzotto et al (2006) investigated water residence times in agricultural areas and found out that residence was shortened from 80 years to less than 40 years due to irrigation. Thus, mobilized arsenic in shallow sediment layers may be transported to the shallow aquifer much faster.
- Jessen et al (2008) developed an adsorption isotherm based on field experiments in Dan Phuong, Vietnam. He postulates that the mobility of arsenic increases rapidly when its concentration exceeds 100µg/l.
- Humic or fulvic acids were also assumed to be a driving factor in As mobilization, because humic acids may form stable complexes with ferric hydroxides and thus compete with As (Bauer, Blodau 2006b) and can also can build stable colloids with As and reduce the mobility of As in the solute phase (Liu et al. 2012). However, (Burton 2013) showed that the presence of humic acids in soil has little effect on the mobilization of As.

2.4.3 As mobilization in paddy fields

Rice cultivation is the most abundant form of agriculture in the Red River Delta. For nine months of the year, the paddies are submerged, and for over 5,000 years mankind has shaped the landscape in order to optimize the interplay of tilling the paddies, flooding and draining after harvest. Plowing, puddling and flooding provoke profound changes in the soil characteristics such that in pedological terms, the paddy soils are considered a separate unit in soil classification). In contrast to other arable soils, oxygen is depleted, partly because oxygen diffusion is much lower in water than in air. Paddy soils are reorganized soils (colluvisols) with a higher level of organic matter, a lower ratio of humic acids to fulvic acid, and the reduced aromaticity of the humic compounds, moreover, its organic matter consists of simple compounds (Peng, Wu 1965). In addition, paddy soils are characterized by eluvation processes: Fe and Mn are dislocated and leached from the plowed horizon and accumulated in the illuvial horizon (Gong 2007), which has a considerable influence on the distribution of As in the upper soils.

Microbial processes are one of the driving factors regarding the specification, mobility and bioavailability of As in paddy soils. Although As is a cell toxin, it is integrated into a range of microbial metabolism pathways, which lead to a change in the As specification such as the reduction of As(V) to As(III) or vice versa the oxidation of As(III) to As(V). The microbial oxidation of As(III) to As(V) is considered a detoxification mechanism (Paez-Espino et al. 2009). Furthermore, the bacterial methylation of As compounds is a common process, leading to a variety of methylated As compounds and to volatilization Yin et al. (2011).

2.5 Arsenic occurrence in daily rural activities

- Review of the related studies

Although rural life in Vietnam is undergoing great transition, most of the daily life activities in the villages still follow traditional pathways. The INHAND project gathered detailed information on mass flow in one work package carried out by the University of Hanover (Meier 2015). Another work package included the planning, construction and operation of a multistep wastewater treatment plant consisting of an SBR unit, a biogas reactor, and a digestate drying facility. For this thesis, some relevant sources and pathways (

Figure 6) have been analyzed regarding their arsenic content.

The characteristics of the key pathways have been investigated in many international studies. A review of these studies is carried out below.

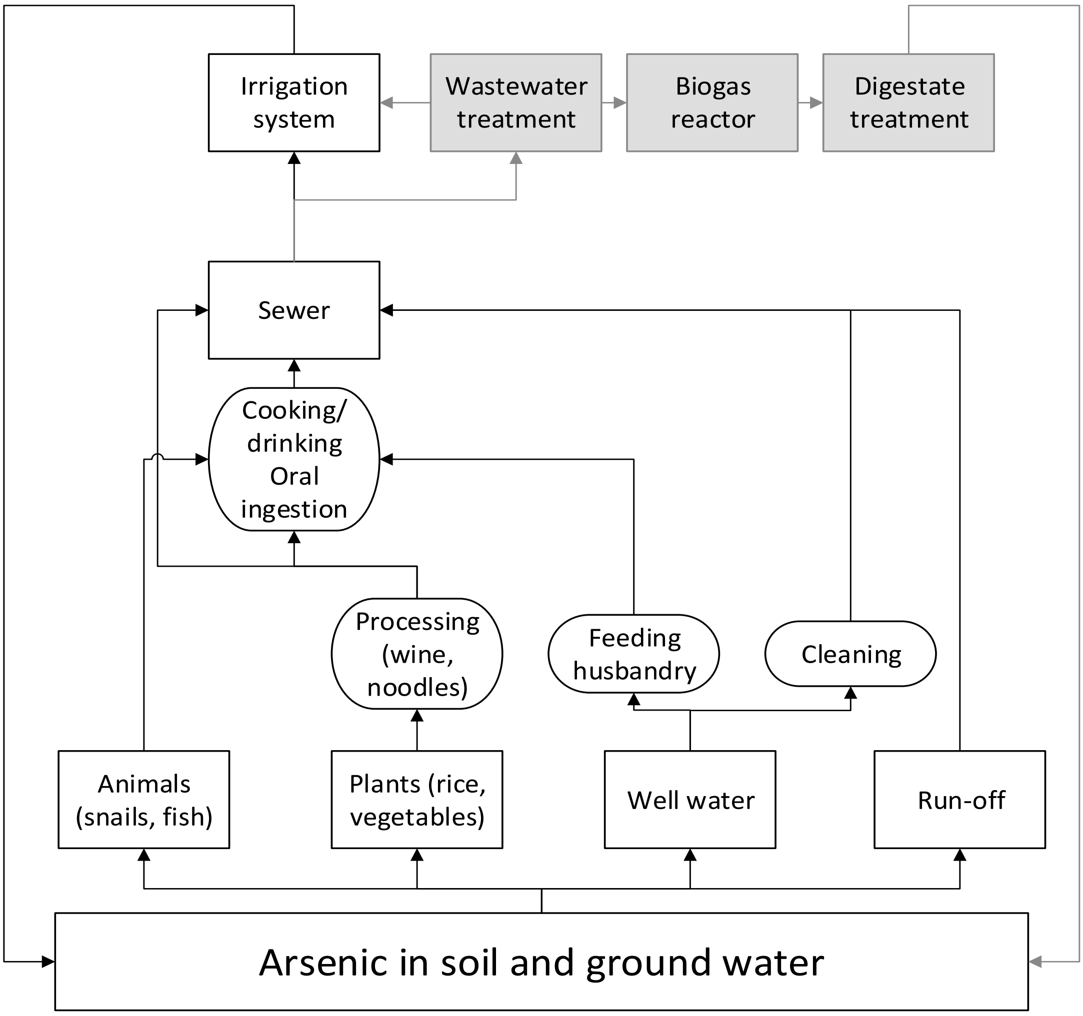

Figure 6: General mass flow of arsenic in a food processing craft village with and without wastewater treatment

2.5.1 Arsenic in soil

The global mean concentration of arsenic in the pedosphere is 5.0 –7.5 mg/kg (Matschullat 2000). Higher values can occur because of human activities such as coal and metal mining, sewage, the use of phosphate fertilizers, wood preservatives and paint, the application of herbicides, or because of natural processes (weathering, sedimentation). One of the most prominent arsenic compounds is cacodylic acid, a herbicide. It was the active ingredient in Agent Blue, which was used during the Vietnam War, when it was an important part of the US government's 'rice-killing operation'. Over 1.2 million gallons of Agent Blue were sprayed on Vietnam's paddies between 1962 and 1971 (Figure 7). No references are available about the degradation of Agent Blue or the possible residual level of Agent Blue in the soil of Vietnam.

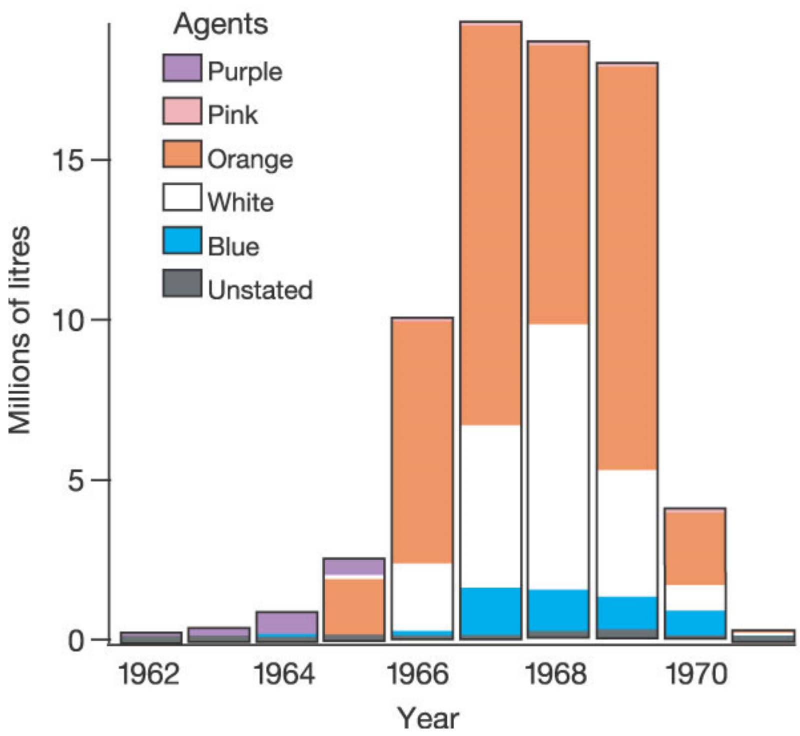

Figure 7: Liters of herbicides sprayed over 1962–1971 (Stellman et al. 2003)

Nevertheless the arsenic concentrations found in the soils of the affected areas in Asia are denoted as geogenic and originate from the Himalayan massif.

The arsenic content in soil is one of the key factors for arsenic uptake in plants and thus for the human and environmental risk. However, even though the indication of the total arsenic content is certainly important, it doesn't reveal the potential risk emanating from soil because the arsenic compounds and species have different toxic and mobile properties.

In most soils, arsenic is mainly available in its inorganic forms like arsenate and arsenite (Cullen 1989), which are regarded as more toxic than the organic components. However, in peatland pore water and forest soils, high ratios of organic arsenic compounds have been found (Huang, Matzner 2007, Huang 2006).

Previous studies in Bangladesh showed generally lower arsenic concentrations in the topsoil (0–15 cm) than in the lower soil (15–30 cm). Meharg (2003) found upper soil concentrations of 3.1 to 42.5 mg/kg. Several studies have shown that there is no significant accumulation of arsenic because of irrigation activities in the upper soil layers over a longer period (Meharg 2003, Lu et al. 2009). Although arsenic accumulation can occur during dry seasons, no long-term accumulation was observed (Sahu 2012, Dittmar et al. 2010). In contrast to Bangladesh, using tubewell water as irrigation water is uncommon in Vietnam because the system is based on the use of river water. In Vietnam, the permissible limit for arsenic in agricultural soils is 12 mg/kg. No comprehensive data pool is available regarding arsenic concentration in the soil in Vietnam. Tran (2002) investigated a number of unpolluted soils, which did not show particularly elevated values. Phuong (2008) found values between less than 5 mg/kg to 42 mg/kg in the soil layer of 20–25 cm.

Regarding the uptake potential of arsenic in plants, the level of mobile arsenic is more significant. A suitable method to assess the mobility of metals in soils is sequential extraction fractionation (SEF). This method assesses trace element pools of differential relative lability in soils. Different reagents of increasing strength are used sequentially and specifically dissolve the bonded phases. The sequential extraction technique used to study the solid-phase association of elements was originally proposed by Tessier (1979) and modified by many researchers (Voigt et al. 1996, Roussel et al. 2000). SEF has been extensively applied to metals and metalloids. The extractants are generally applied in the following order: unbuffered salts, weak acids, reducing agents, oxidizing agents and strong acids.

The common fractions of As extraction consist of 1. Desorption of slightly (nonspecifically and specifically sorbed) phase, 2. Desorption of Fe-, Al- or Mn-oxyhydroxide, and 3. Residual phase.

Besides, there are other operationally defined extracted fractions such as water-soluble or easily soluble, acid sulfide, bound to organic matter, acid-soluble, bound to carbonate, arsenic oxide and silicate, and arsenic or iron sulfide. The extraction chemicals in arsenic SEFs differ depending on the study's aim. For example, the fraction of exchangeable As is normally extracted based on the principle of ion exchange, i.e. the loosely bound arsenic is exchanged with one of the constituents of the extraction chemicals. To extract this fraction, (Tessier) 1979 used $MgCl_2$ while Wenzel et al. (2001) used $(NH_4)_2SO_4$. Another example is the strongly adsorbed As fraction, which is often extracted by phosphate salts because of the competitive exchange between phosphate (PO_4^{3-}) and arsenate (AsO_4^{3-}) in soil (Hudson-Edwards et al. 2004). In order to extract this fraction, Keon et al. (2001) and Wenzel et al. (2001) used $NaHPO_3$ and NH_4PO_4 respectively.

In this study, an SEF method adapted and approved by Wenzel et al. (2001) was chosen to investigate the fractionation of the As pool because of its good applicability and rate of recovery.

2.5.1.1 International standards for As in soil

The international standards for As in soils provide a wide range of thresholds, reflecting the complexity of classification. German legislation (Tessier 1979, Guo et al. 2003) states a test value of 50 mg/kg in residential areas, 25 mg/kg in playgrounds, and 125 mg/kg for parks and recreation areas. In grassland soils, the test value is 50 mg/kg and in agricultural soils 200 mg/kg, although under reducing conditions the threshold is 50 mg/kg.

In 2009, the Environmental Agency of Great Britain issued guidelines to handle As in soils. In residential areas, the maximum value is 32 mg/kg, in garden areas 43 mg/kg, and in commercial land 640 mg/kg.

In Vietnam, the national standard for As in agricultural soils is 12 mg/kg (Phuong 2008)

2.5.2 Arsenic in drinking water

The occurrence of As in shallow groundwater in the Red River Delta was detected more than fifteen years ago. The origin and mobilization of As in the Red River Delta was described in chapter 2.4.

Removing As from groundwater to reduce the concentration to below international standards isn’t a technological problem: ion exchange, activated alumina, reverse osmosis, membrane filtration, modified coagulation/filtration, and enhanced lime softening are all recommended treatment technologies for As removal (US EPA 2001). However, these technologies are hardly applicable in rural Vietnam due to the costs and lack of technical resources. Many households in the Red River Delta use simple sand filters to remove Fe from drinking water. After the water has been pumped up from the groundwater, it passes through a sand filter and is stored in a basin (Berg et al. 2006) (see Figure 8). The original purpose of the sand filters was to reduce the content of Fe, which has a ‘nasty taste’ in the water due to oxidation and precipitation/adsorption. The adsorbed Fe(II) and oxidized Fe(III) leads to the oxidation of As(III) to As(V), which is less mobile and less toxic. Berg et al. (2006) proved the diminution of affected drinking water samples to less than 50 µg/l in 90% of the samples.

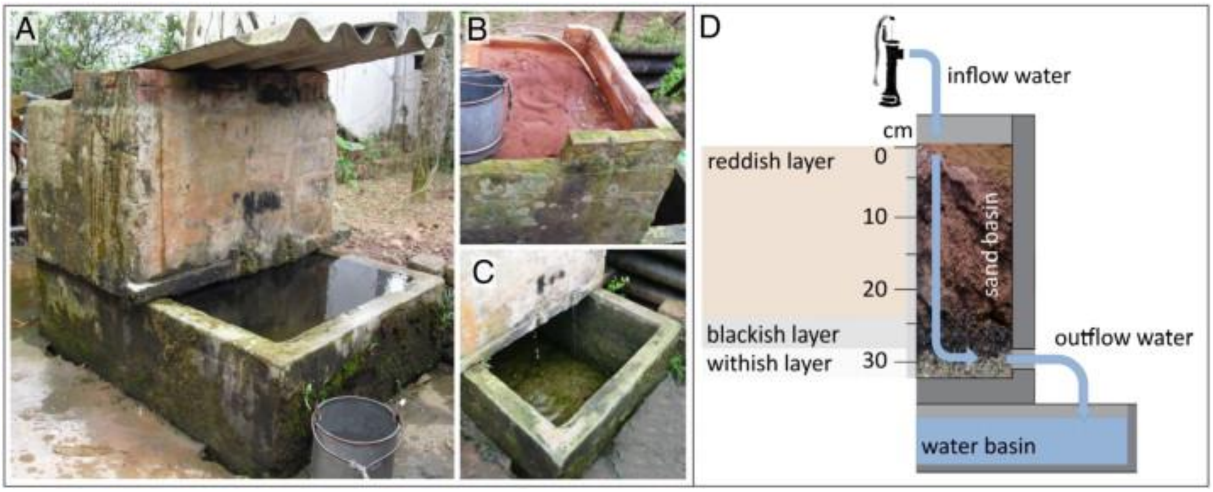

Figure 8: Household sand filter consisting of two open containers made out of concrete or brick. The upper container covered with a corrugated metal sheet serves as a filter and the underlying tank is used to store treated water. The upper container must have one or more outlets either at the bottom (2) or in the front wall (3). A simple sieve (e.g. a piece of cloth) is used to prevent the sand from flushing out of the filter. The valve (5) serves to empty the storage tank for cleaning (Berg et al. 2006)

2.5.3 Phytoaccumulation: Current state of research

The phytoaccumulation of arsenic has been studied for more than seventy years. Early studies from the 1940s dealt with the phytotoxic effects of arsenic uptake on crops following its use as a herbicide and pesticide (Fleming et al., Jones, Hatch S. H. 1945). New analytical methods for the detection of arsenic species provided a closer insight into the complexity of arsenic species in plants (Braman, Foreback 1973). This research was given a boost at the turn of the millennium when the poisoning catastrophe in Bangladesh and West Bengal became increasingly obvious.

Throughout the last decade, special scientific interest was been directed at the dynamics and potential of arsenic uptake in rice. From 2007 to 2014, nearly 700 articles on arsenic in rice were published in international scientific journals. From 2002 to 2006, 156 articles were published in the same field, while from 1900 to 2001, the number of related articles was just about 30 (Figure 9) (Thomson Reuters 2014). Even when we take into account that the number of scientific publications generally increased significantly in the same period, these numbers indicate the growing interest and concern in the problem of arsenic contamination in rice.

The sharp rise of interest in arsenic uptake in rice is explained by the growing concern regarding groundwater contamination in Bangladesh, West Bengal and Vietnam in the 1990s. Rice is a staple food for about half the world’s population (FAO 2013): over 740 million tons were produced worldwide in 2013 and production was forecast to grow by 0.8 percent in the following year (FAO 2014). Much of this rice is grown in areas affected by arsenic contamination (the Bengal Basin, Irrawaddy Delta, Mekong Valley, Red River Delta, Indus Plain, Yellow River Plain). Hence, the problem of contaminated rice is not limited to the distinct regions – for Asian rice is exported throughout the world. Accordingly, the numbers of scientific studies on arsenic uptake increased strikingly in 2002. Most publications are written in China, followed by the USA, India and Bangladesh. In 2007, another abrupt rise in the numbers of publications was noted from around 30 to more than 80 publications annually, underlining its relevance.

In 2002, a Scottish team of researchers headed by Andrew Meharg studied arsenic uptake in rice in detail, and four important publications about arsenic uptake in rice from Abedin (Abedin et al. 2002b, Abedin 2002a, Abedin 2002b, Abedin et al. 2002a) launched a whole raft of publications. Abedin et al. studied the kinetics, accumulation and metabolism of arsenic in rice plants. They proved that 95% of the arsenic in rice shoots is inorganic arsenic and a large share is arsenite; they also found arsenite to be the species with the highest concentration in paddy fields, creating a serious health risk (Abedin 2002a).

In the last decade, Meharg's team has studied different aspects of arsenic uptake in rice plants. More than 70 publications have provided important insights into the uptake kinetics and mitigation. Meanwhile, the most active researcher group in the PR of China is Yonguan Zhu from the Chinese Academy of Sciences in Beijing. A soil scientist, Zhu et al. focused on soil characteristics and preconditions for arsenic uptake in rice.

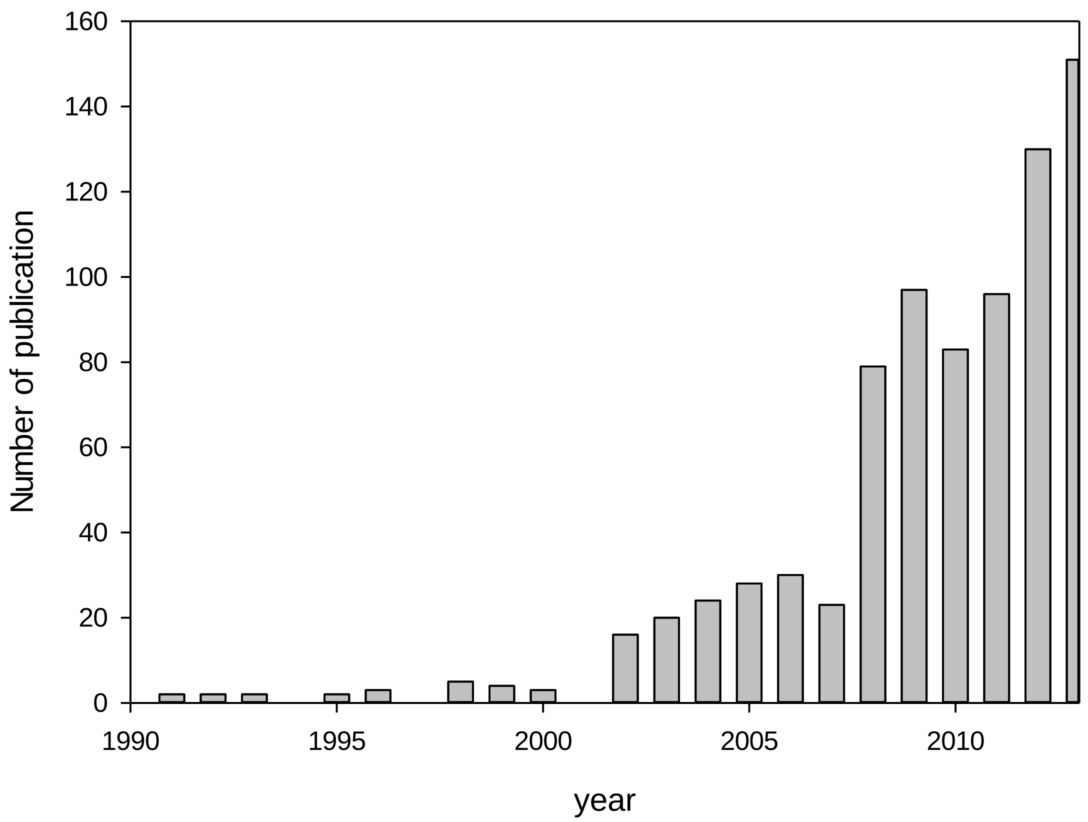

Figure 9: Number of international articles related to arsenic in rice

2.5.4 Bioavailablity

The mean global arsenic concentration in soil is 5.0-7.5 g mg/kg. However, the uptake of arsenic in plants depends on the availability of arsenic in soil water. To some extent, arsenic is embedded in soil minerals or is adsorbed strongly on the mineral surface, and iron plaques play an important role as adsorbent for arsenic in soils. The specification of arsenic as well as pH, redox conditions, oxygen, microbial activity, the soil temperature and organic content have a strong influence on the mobilization and specification of arsenic, which are the key properties for bioavailability.

There are several reasons why plants take up arsenic, even though this may have adverse effects.

1. As and P are both members of the nitrogen group of the periodic table and have some clear similarities. The oxides arsenate and phosphate also display some commonalities in their physicochemical behavior. Phosphate, an important anion for plants, thus competes with arsenate (Lei Ming 2014).

2. Arsine enters plant cells through nodulin 26-like intrinsic protein (NIP), an aquaporin transporter which is normally provided to transport small molecules, glycerol and water. Ma et al. (2007) proved that arsinic acid, which has similarities to silicic acid, enters the rice roots via aquaporin transporters.

Plants can take up inorganic and organic arsenic species. Due to their different sorption and solubility properties, the availability of the species varies significantly. In soils which are not subject to anthropogenic pollution because of herbicide application, the inorganic species As(III) and As(V) are the predominant species. In aerobic soils, As(V) is the most abundant species, although its bioavailability is restricted due to the strong tendency to adsorb on iron(hydr)oxides. As(III) predominates in anaerobic soils such as in submerged paddy fields because the conditions evoke a reductive dissolution of the iron(hydr)oxides and hence the release of adsorbed arsenic compounds. Furthermore, As(V) is reduced to As(III), which is more mobile. In aerobic soils with low pH values, As(V) is easily interconverted to As(III) (F). Microbiological activity is important for reduction, oxidation and methylation (Fendorf 2008, Zheng 2013, Huang 2011).

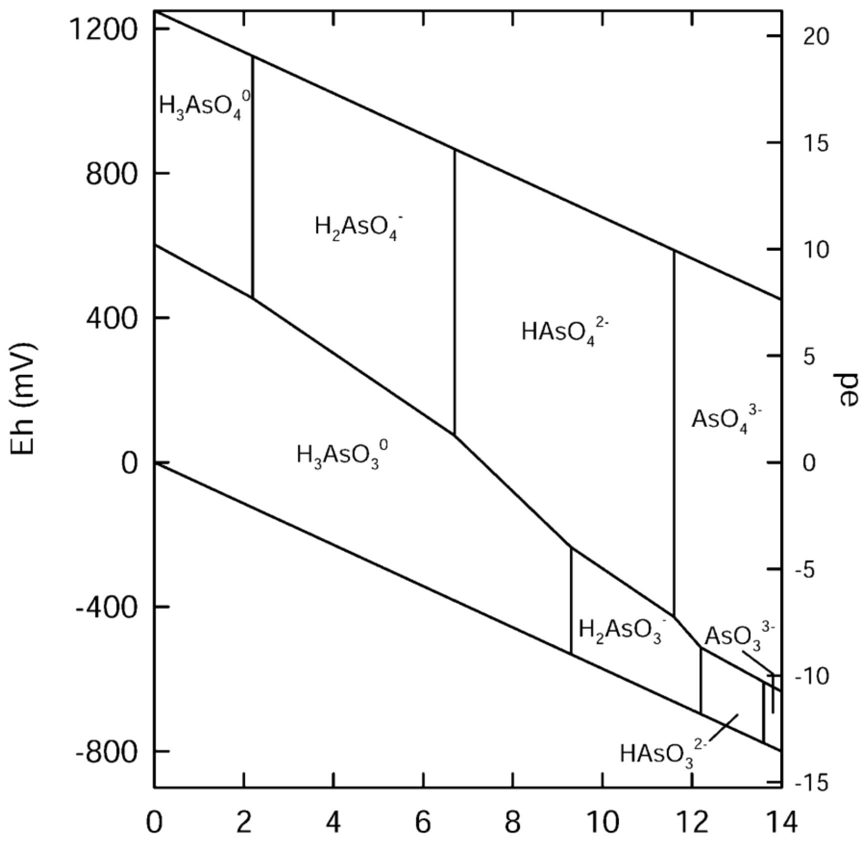

Figure 10: Eh-pH diagram for aqueous As species in the system As–O_2–H_2O at 25°C and 1 bar total pressure (Smedley 2002)

Weathering is an important regulating factor for the bioavailability of heavy metals because earth-alkaline metals are leached as carbonates and sulfates; Si, Fe and Al are more immobile and can form oxy-hydroxides which are adsorption partners for heavy metals. Thus, the bioavailability of As is not affected by weathering processes (Chang et al. 2014).

Organic arsenic compounds are commonly found in soils. MMA and DMA can be formed from As_{in} by methylation through microorganisms. DMA is the predominant organic species, especially under anaerobic conditions and when the organic matter is relatively high (Huang 2011). All organic species can be taken up by plants and can be detected in the plants; however, studies of the uptake of methylated arsenic compounds are scarce. Jedynak (2010) proved the accumulation of DMA and MMA in white mustard plants and showed that both species are transferred to other methylated arsenic species and into $As_{in,}$ whereas plants are not able to methylate As_{in}.

2.5.5 Arsenic uptake in rice plants

The fact that rice plants are able to accumulate more arsenic than other crops can be explained by the rice plants' protective mechanism. Rice is a typical silicon-accumulating plant and can accumulate up to 10% silicon in the shoots, which is required for robust growth and resistance to pest attack, fungal infection (Rémus-Borel et al. 2005), and other biotic and abiotic stresses (Ma, Yamaji 2006). In shoots, stems and leaves, silicon forms small bodies (Ma et al. 2007). In flooded paddies, silicon and arsenic are chemically similar and arsenic fits into the silicon transporters, which can lead to arsenic accumulation in all compartments of the plant. The availability of silicon in soils correlates negatively with arsenic concentration in rice plants (Bogdan, Schenk 2008).

The rhizosphere of rice plants grown in paddy fields differs from that of other crop plants. Many rice species are marshplants with the ability to transport oxygen from the aerated parts of the plant to the aerenchymes. This leads to oxidizing conditions in the rhizosphere known as radial oxygen loss (ROL) (Colmer 2003). In addition to the redox conditions, the pH is reduced significantly (Pan et al. 2014). These conditions can increase the mobility of some trace elements and also lead to the oxidization of iron or manganese and thus to the formation of iron oxides and iron hydroxide plaques on the roots (Brouwere 2004). It was shown that flooding leads to the mobilization of arsenic (Li et al. 2009). Pan et al (2014) proved that there is significant variation in arsenic specification in the rhizosphere during the flooding period and that the concentration of mobilized arsenic is slightly higher in the rhizosphere soil than in the surrounding soils. Different genotypes of rice show different development of ROL and hence different abilities to take up arsenic. In addition, the specification of the accumulated arsenic in the plant differs from one genotype to the next (Wu et al. 2011).

2.5.5.1 Arsenic uptake in vegetables

Food vegetables like carrots, radishes, tomatoes and spinach take up arsenic in proportion to the arsenic concentration in the soil or irrigation water, although distribution in the plant is not homogeneous. Radishes and tomatoes accumulate higher levels in the leaves and skins than in the roots; carrots have higher concentrations in the roots than in the aerial parts. Like most other plants, As uptake in leaf vegetables occurs via the roots. The few studies of arsenic uptake in leaf vegetables demonstrate that the ratio of arsenic uptake in leaf vegetables is higher than other heavy metals like Hg, Cd Pb and Cr (McBride 2013, Chang et al. 2014). Spinach shows the highest ability for arsenic uptake (Bhatti et al. 2013). Farid (2003) showed that leafy vegetables accumulate higher levels of As in aerial parts (edible shoots) than other vegetables. In contrast to other heavy metals which probably enter leaf vegetables by atmospheric deposition (Pandey, Pandey 2009), As is taken up via the root pathway (Mc Bride, 2013). In some studies, the concentration in the plant clearly correlates to the content of arsenic in the irrigating water (Baig, Kazi 2012). One study carried out in Bangladesh in 2004 (Das et al. 2004) detected elevated concentrations in arum leaves (*Colocasia antiquorum*), a common tropical-subtropical leaf vegetable (0.09-3.99 mg/kg), in potatoes and in *Ipomoea reptans*, also known as water spinach, leafy vegetable kalmi and morning glory (0.1–1.53 mg/kg). This vegetable is grown and eaten in many regions in Asia, its leaves and stems contain many vitamins and nutrients, and it is grown in wetlands, where anaerobic conditions may increase As uptake. Shaibur (2009) carried out greenhouse experiments with *ipomoea reptans* and different As concentrations in the irrigation water. He found that *ipomoea reptans* showed almost no toxic response even at very high As concentrations and that the accumulation of As followed the trend: root > stem > leaf.

The EFSA report nevertheless includes 1,903 samples of leafy vegetables. Within these data, the leaves and sprouts of *Brassica spp* had the highest arsenic content with 46.3 µg/kg. The mean of the spinach samples is 11.9 µg/kg and the mean of 546 samples of unspecified leafy vegetables is 8 µg/kg.

2.5.5.2 Arsenic uptake in rice

In contrast to other crops, some rice species have a high potential to accumulate arsenic in the shoots and the grains. Many studies have proved high arsenic concentrations in rice from all over the world. Meharg (2003) reported that rice grains collected in contaminated soils in Bangladesh had concentrations that were ten times higher than the average concentration of 0.1 mg/kg. (Williams et al. 2005) described high arsenic concentrations in rice from the USA resulting from the application of monosodium methylarsenate (MSMA) and disodium methylarsenate (DSMA) as herbicides on cotton fields for decades. In contrast to the affected rice samples from Bangladesh and India, it was shown that the US rice samples had higher concentrations of organic arsenic components than the Asian samples, indicating a lower health risk (Williams et al. 2005).

The As concentration in rice from Bengal, Mekong, Irrawadi or Red River deltas varies enormously (Table 2). Global As concentration in rice samples ranges between 0.02 and 0.8 mg/kg dry weight (Zavala, Duxbury 2008).

Table 2: As concentrations in rice in international studies [mg/kg]

	Mean	Min – max	n	
Bangladesh	0.13	0.1-0.95	175	Williams (2005)
	0.14		10	Das et al (2004)
		0.08-0.34		Dittmar et al. (2010)
		0.07–0.74		Norton et al. (2010)
		0.192–0.899		Norton et al. (2012)
		0.1–0.72		Stroud et al (2011)
West Bengal	0.13	0.02-0.17	50	Modal et al. (2008)
		0.19-0.78		Bhattacharya et al. (2010)
		0.05–0.84		Norton et al. (2009)
				Stroud et al.,(2011)
China	0.5	0.31-0.93	11	Williams (2006)
	0.09			Zhu et al. (2008)
	0.09	0.02-0.18		Jiang et al. (2014)
		0.01-0.31		Fu et al. (2011)
		0.31-0.52		Lei et al. (2013)
		0.07-0.27		Liang et al. (2010)
		0.27–0.85		Norton et al. (2009)
Taiwan	0.1	<0.1-0.63	426	Williams (2006)
USA	0.26	0.11-0.46	18	Williams (2006)
	0.09	0.04-0.144	284	WHO (2012)
Europe	0.18	0.13-0.22	7	Williams (2006)
		0.07-0.4		Sommella et al. (2013)
		0.1	706	EFSA (2014)
Vietnam	0.21	0.03-0.47	31	Williams (2006)
		0.29-0.66		Hsu et al. (2012)
		0.093–0.345		Nookabkaew et al. (2013b)
Cambodia	0.31		14	O'Neill et al (2008)

*As_{in}

2.5.6 Arsenic in meat and animal products

The presence of As in meat and animal products has come to the public eye because of the use of organic arsenicals as pesticides (roxarsone) in poultry production in the USA and because of the risk that As can enter the human food chain. Even if As is accumulated in the feathers and not in the meat of poultry, feather material is often reused as fertilizer or as feather meal in other livestock breeding (Nachman et al. 2013). However, in Vietnam the use of organic arsenicals is not documented. In fact, chickens in the villages are fed waste and residue from piggeries, and so the potential sources of As in chicken are rice grains, worms, insects, household waste and dust. As analyses of poultry products in Vietnam have yet to be published.

Very few studies have published results from As analysis in pork meat and pork liver (Table 3). Bordajandi et al. (2004) found a mean concentration of 0.0624 in pork meat samples in Spain. In Sweden (Jorhem et al. 1991), the mean As concentration in pork meat was 0.024 ± 0.023mg/kg(ww) and in pork liver 0.023 ± 0.022 mg/kg (ww). Food sample analyses in Thailand revealed a higher As concentration in meat samples than in European samples (Nookabkaew et al. 2013a). Pork meat samples ranged between < 0.0016 and 0.189 mg/kg and pork liver samples even had a concentration between < 0.0016 and 1.087 mg/kg (mean 0.131 mg/kg).

Table 3: As in animal products

Study	Concentration [mg/kg]	Country
Chicken meat		
(Gagnon et al. 2004)	0.33–0.43	USA
(Bordajandi et al. 2004)	n.d.–0.178	Spain
(Leblanc et al. 2005)	0.022	France
(Ishizaki 1979)	0.017–0.033	Japan
(Rana 2014)	0.385 + 0.125	India
(Nachman et al. 2013)	0.0026–0.0036	USA
Chicken liver		
(Bordajandi et al. 2004)	0.315	Spain
(Korsrud et al. 1985)	<0.07–1.5	Canada
(Gutu 2011)	0.405 ± 0.015	Romania
Eggs		
(Bordajandi et al. 2004)	0.033–0.04	Spain
(Daghir, Hariri 1977)	0.002–0.240	Canada
(Leblanc et al. 2005)	0.008	France
(Munoz et al. 2005)	0.019	Chile
(Ishizaki 1979)	0.028–0.034	Japan
(Horiguchi et al. 1978)	Trace–0.097	Japan
(Rana 2014)	0.222 + 0.120a	India

2.5.7 Arsenic uptake in golden apple snails

The golden apple snail (GAS) (*Pomacea canaliculata)* is an aquatic gastropod mollusk with gills and operculum and is able to respire in water and in air (Mochida 1991). The species is widespread in paddy rice fields in Asia and often serves as a food source.

Its direct contact to soil, soil water and irrigation water begs the question of whether the GAS accumulates arsenic.

Originally the GAS was introduced in 1979 from the tropical regions of South America to Taiwan. It was supposed to be raised as a delicate food in Asia, but just three years later GAS was reported as a pest in Taiwan in 1982 (Cheng 1989) and in most other Asian countries in the following years (Litsinger J.A, . Estano 1993). The lack of natural predators and optimal progeny and living conditions led to high abundance and spatial distribution. GAS often pose a threat to rice plants and subsequently the rice harvest in East and Southeast Asia. The economic damage is significant (over US$40 million in the Philippines ten years ago) (Hayes 2008). The application of pesticides is problematic and cost intensive. Although the use of the GAS as a food source was not very common for a certain period, GAS are now an inherent part of many Vietnamese menus. However, their use as a nutrient source also has a number of drawbacks. GAS are an alternate host of the parasite *Angiostrongylus cantonensis (*rat lung worm), which causes eosinophilic meningitis, a serious condition that can lead to death or permanent brain and nerve damage (Li et al. 2008). In addition, mollusks are known to be able to accumulate large quantities of heavy metals (Deng et al. 2008). Even so, only few studies have investigated arsenic uptake in freshwater snails. Lai et al. (2012) conducted a study on freshwater snails in Thailand in which he addressed the specification of arsenic in two different snail species. The researchers found up to 83 mg/kg (dw) in the snails; the main species were TETRA and arsenosugars.Hong et al. (2014) analyzed different aquatic species in a highly industrialized city in Korea. It was found that pond snails contained up to 6.4 mg/kg As (dw). In contrast to mollusks, shrimps, bivalves and fish, the gastopodes contained only a small amount of the harmless arsenobetaine but a high level of inorganic trivalent As. In Vietnam, where the consumption of GAS is becoming more and more popular and where arsenic in paddies is a known threat, no information on arsenic or heavy metal content in GAS is available.

2.5.8 Processing: Wine and noodles

The village of Dai Lam is one of the 3,500 traditional craft villages in Vietnam, which represent an important pillar of the Vietnamese economy. The traditional craft of Dai Lam is to produce rice noodles and wine from cassava and rice. The material flow induced by the processing activities may play a relevant role in assessing the As exposure of the population and can be transferred to other food processing villages:

The rice processed in Dai Lam is grown in the area of the village; the cassava is grown elsewhere and transported to Dai Lam. The main processing water used is tubewell water, which contains heavy metals and As in varying concentrations. Additionally, in many food processing craft villages, pigs are raised in the producing households in order to utilize the residue from wine processing (slurry). The pigs are either slaughtered in Dai Lam or transported to marketplaces in Bac Ninh. The piggeries produce a substantial amount of manure, which is often used as fertilizer in gardens or sold to traders who deliver the manure to other gardening companies in the province; alternatively, the manure is flushed into the sanitation system and transported untreated to the paddies or receiving streams.

No scientific studies have been published yet which examine the relationship between heavy metal and As exposure and processing activities in craft villages.

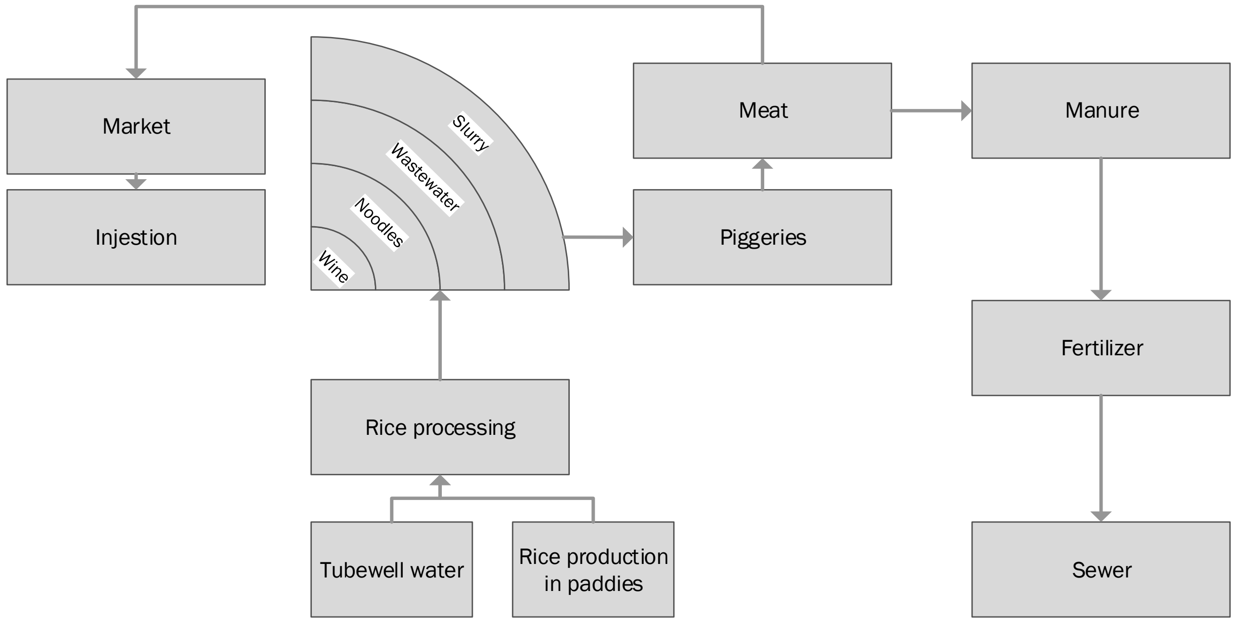

Figure 11: Relevant material flow steps of rice processing

2.5.9 Arsenic concentrations in wastewater, activated sludge and digestate

In the study area, a three-step pilot plant was designed, built, and put into operation during the research project INHAND. The aim was to investigate the mass flow and treatment efficiency of an SBR. In addition, a biogas reactor and a digestate drying facility were installed.

Only a few studies have investigated the behavior of arsenic in biological wastewater treatment processes. Goldstone (1990) found arsenic's characteristics in wastewater to differ from those of other heavy metals, the activated sludge process in a full-scale treatment plant only removing 34% of the As; similar results were reported by Watanabe et al. (2002). The SBR process is one of the most common activated sludge processes to be used for wastewater treatment. Andrianisa et al. (2006) studied the behavior of arsenic species in SBR batch tests and noted that the aerobic treatment process oxidizes As(III) to As(V). MMA was simultaneously methylated to DMA and further to inorganic As(III), which was subsequently oxidized to As(V). Though the transformation processes occurred within a matter of hours, the treatment efficiency was low. Further treatment with $FeCl_3$ achieved a high arsenic removal efficiency (>95%) by coagulation (Andrianisa et al. 2008).

The anaerobic digestion of sewerage sludge might favor the formation of volatile derivates of arsenic like arsine AsH_3 and mono, di and trimethylarsine: $AsH_2(CH_3)$,

$AsH(CH_3)_2$ and $As(CH_3)_3$ (Michalke 2000, Mestrot, 2013). The production of the volatile compounds is driven by microorganisms, is linked to the reducing conditions, and depends on the reactant and on the availability of hydrogen (Mestrot, 2013).

2.6 Iron and manganese in the nutrient chain

Mn is one of the most abundant elements in the crust and has 11 oxidation states. In nature, +II, +IV and +VII are the most important states. Mn is an essential element for humans and animals, but in higher concentrations may pose a serious health risk. For instance, it accumulates in the brain and in the central nervous system and causes neurological damage (Rodriguez-Barranco et al. 2013). Children appear to be more severely affected by manganese exposure than adults. Several authors have detected a positive correlation between attention deficit hyperactive disorder (ADHD) and manganese exposure (Farias et al. 2010, Ericson et al. 2007, Yousef et al. 2011). A detailed study which investigated the effects of Mn in drinking water in Bangladesh showed a clear neurotoxicological effect on children exposed to drinking water with Mn concentrations above 300 µg/L (Khan 2011). The WHO guidelines for drinking water quote a standard for drinking water of 0.4 mg/l.

In food the concentration of manganese depends strong on the kind of food and on cultivation circumstances (Table 4).

Table 4: Manganese concentration (mg/kg) in food sources: ATSDR (2000)

Nuts and nut products	18.21–46.83	Vegetables and vegetable products	0.42–6.64
Grains and grain products	0.42–40.70	Meat, poultry, fish and eggs	0.10–3.99
Legumes	2.24–6.73	Infant foods	0.17–4.83
Fruits	0.20–10.38	Beverages (including tea)	0.00–2.09

In the WHO drinking water guidelines (background document), the mean Mn intake in western diets is stated to be 0.7 to 10.9 mg/day (Greger 1999, EPA 2004). The NOAEL (non-observed adverse effect level) is 11 mg/day and the tolerable daily intake (TDI) was calculated by dividing the NOAEL of 11 mg/day by 3 (which considers the uncertain increased bioavailability of manganese from water) and an adult body weight of 60 kg. Thus, the TDI is 0.06 mg/kg b.w. The guideline value of 0.4 mg/l is calculated from the TDI by assuming an allocation of 20% of the TDI to drinking water and consumption of 2 liters of drinking water per day by a 60 kg adult.

The presence of As in soil and groundwater is often associated with Mn and Fe. Winkel et al. (2011) investigated the distribution of several metals and other parameters in 512 tubewells in the Red River Delta and provided a good overview of the current situation in the shallow Holocene aquifer (Figure 12). The mean concentration of Mn in the sampled wells was 0.83 mg/L and 44% exceeded the WHO standard of 0.4 mg/L. The mean concentration of Fe in the sampled tubewells was 10.83 mg/L. In the studies mentioned here, no correlation was observed between As, Fe and Mn. In the present study, a positive correlation was observed between As and Mn in some data sets. In this regard, the concentration of Mn and its relevance to health risks is mentioned alongside As in the following chapters.

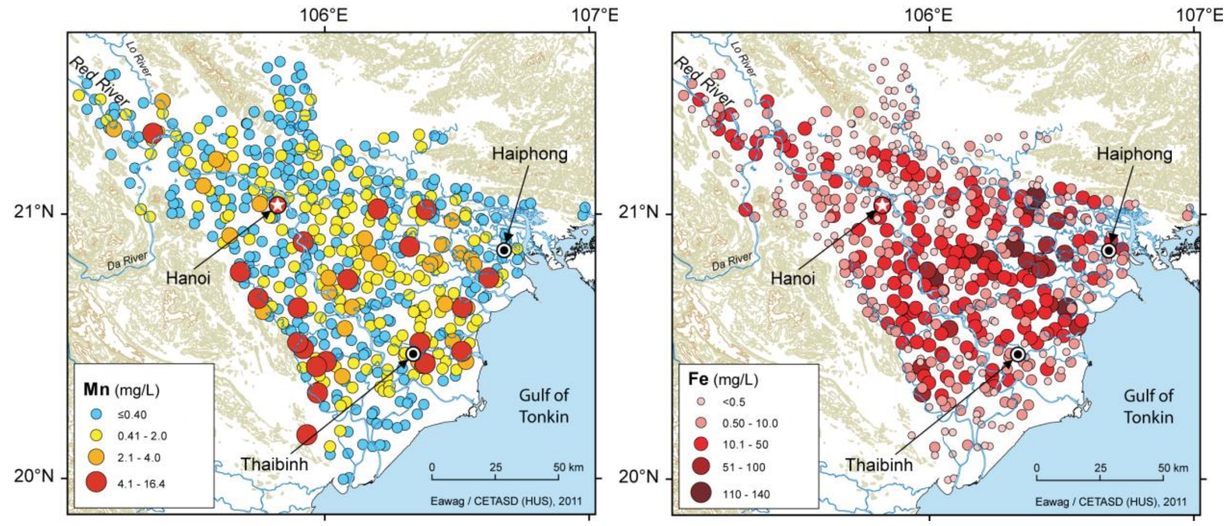

Figure 12: Manganese and iron in tubewell samples in the Red River Delta (Winkel et al, 2010) Supplementary information to research article „arsenic pollution of groundwater in Vietnam exacerbated by deep aquifer exploitation for more than a century" published in PNAS. Doi. 10.1073/pnas 1011915108 Weblink: www.eawag.ch/arsenic-vi-etnam

2.7 Land and water use in the Red River Delta

Land and water use in Vietnam will undergo crucial changes over the next fifteen years in response to both global and national issues Figure 13).

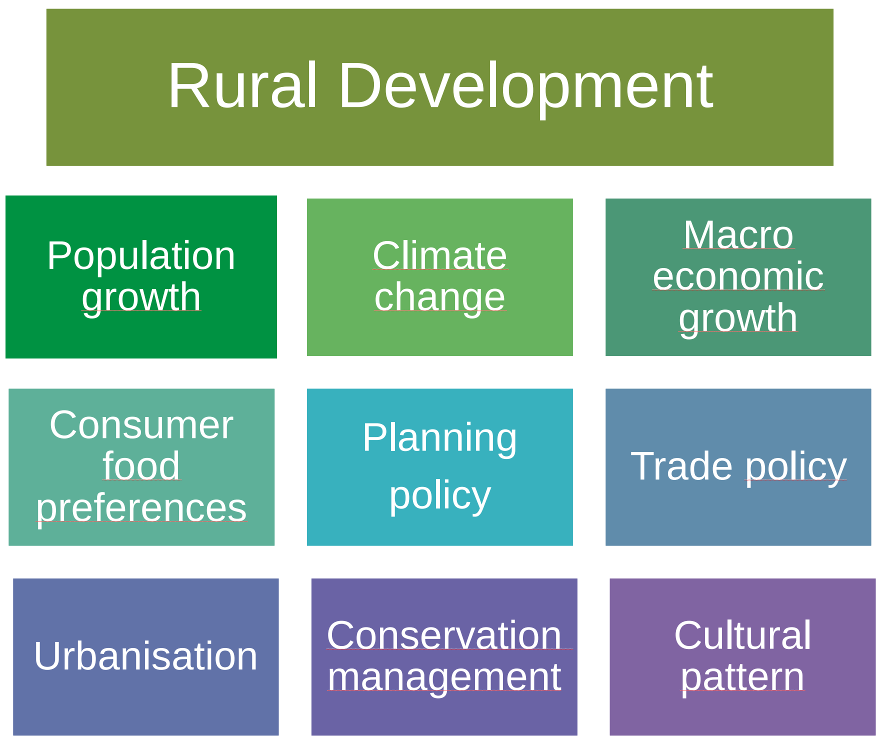

Figure 13: Driving forces for rural development

In 2030, the population will number 104 million, meaning it will have doubled within fifty years. After 2030, population growth will slow down, the *ifs* predicting a population of 108 million in 2060. The highest population growth will take place in the Mekong Delta and the Red River Delta. The urbanization of Vietnam accounts for 32.2% today and will increase to 44.5% in 2030 and 65.77% in 2060.

"Viet Nam is likely to be one of the countries most vulnerable to climate change and is likely to be significantly impacted by it." (IPCC, 2007). This vulnerability is explained by different facts: more than half of Vietnamese live in the Delta regions with an increasing and direct risk of typhoons and floods. Several scenarios (from low impact to worst case) are presented in several studies. It is likely that in north Vietnam the temperature will increase by several degrees, the numbers of days with an average temperature of 25°C rising from 124 days to 176 days in 2005 and 204 days in 2010. The rising temperature will increase the crop rate and shorten the plant growing cycle. However, the demand for agricultural water may double or triple in 2100 compared to 2010. Also, the runoff of the two major rivers in Vietnam will change substantially, decreasing during droughts but rising in the humid season. As a result, the output of the spring rice crops will drop by 3.7% by 2020, 12.5% by 2050, and 16.5% by 2070. The winter crop will drop by 3.7 by 2050 and 5. by 2070 (FAO 2011, IPCC 2007).

Vietnam has made some impressive and effective improvements in poverty reduction. Today, the GDP per person is stated to be US$ 5,070 and is set to increase to US$ 8,540 in 2030 and US$ 21,690 in 2060. Although this economic growth is part of its transition to a highly developed country, these changes are associated with changing consumer behavior which will have important consequences for agriculture and thus for water and land use.

Vietnam is one of the fastest growing economies in Southeast Asia and is listed as one of the 'Next Eleven' (N-11) countries which are believed to have a high potential to be one of the largest economies in the twenty-first century (O'Neill et al. 2005). Due to the increase in average income, consumer food preferences are likely to change in the coming decades. The consumption of meat will increase, making a change in animal husbandry from domestic livestock to industrial meat production inevitable. This will lead to an increased demand for crops like soya and maize. The increasing population and growing income will hence result in growing yields.

Regarding the treatment of the rural areas, the People's Committee promulgated Resolution No. 26-NQ/TW on 'agriculture, farmers and rural areas' (called Tam Nong) in August 2008. This resolution forms the basis of all further discussions and decisions. It states the need for a fundamental change, the restructuring and reorganization of the planning of rural development by clear guidelines and targets. The intentions of the Resolution are to improve spiritual and material life in rural municipalities and to set action targets for adaptation to urban circumstances. The National Target Program on New Rural Development (NTP-NRD) represents the framework for all actions in 2011–2020. One of the main goals is to develop and modernize rural areas by coordinating development in agriculture, land use, employment, enterprise and industry, education, health, tourism, infrastructure, environmental protection and the sustainable use of natural resources at municipality level (land, water resources, forest) (FAO, 2013).

The NTP-NRD is a comprehensive institutional program which in practice has turned out to be difficult to implement due to the ‘bunker mentality’ of the institutions involved (Rudengren et al. 2012).

Trade policy has played an outstanding role in Vietnam’s rural development. For example, the *Doi Moi* Policy has opened up the markets and within two decades the productivity of the rural economy has grown strongly. This development also brought about vast changes to land and water use patterns.

The above-mentioned economic changes are accompanied by rapid urbanization, which can be seen in the population numbers of the two core and periphery urban systems as well as in the spatial redistribution of the rural areas. Rapid urbanization is impacting energy consumption, economic development, natural resources, water and land use, the availability of arable land, and human well-being (McDonald et al. 2014, Brown et al. 2009). Urban centers concentrate the water demand of millions of people within a relatively small area. The fast growth of cities in developing countries is accompanied by increasing water demand, and as most of the fast-growing cities are located in developing countries, the lack of financial support for a sustainable water supply inhibits balanced water withdrawal (McDonald et al. 2014).

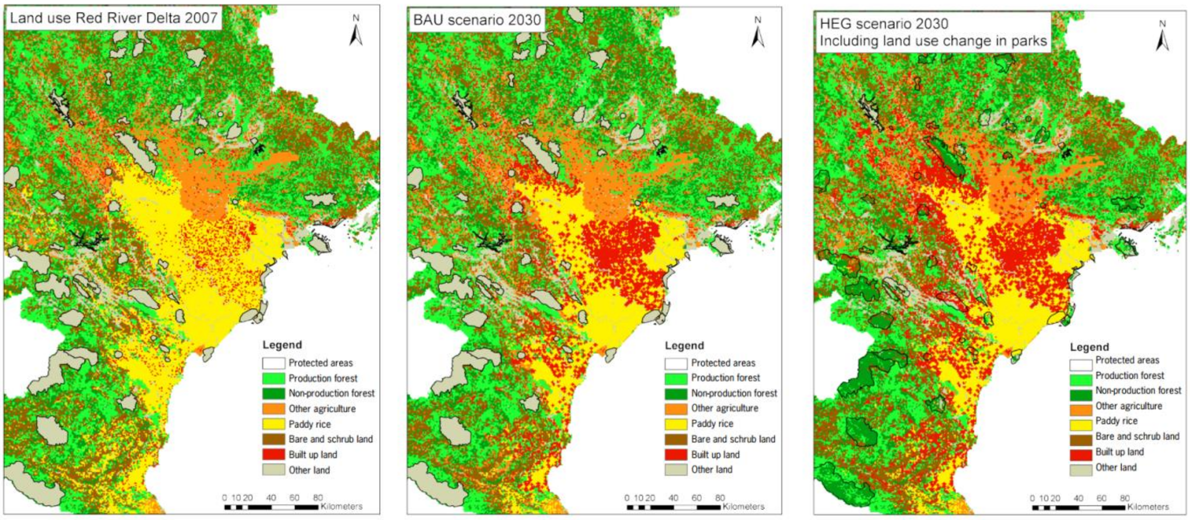

Figure 14: Land use patterns in 2010 and in 2030 (business as usual and high economic growth scenario) (Rutten et al. 2012)

2.7.1 Historical and political aspects of rural development in Vietnam

Land policy and its effect on the historical upheavals have played an important role in the history of Vietnam. The unequal distribution of communal land in the villages was ultimately the main reason prompting the rural population to participate in the revolution. Collectivization in the north from the 1950s and the south in the 1970s didn’t bring about the expected effect for national food security. Staples such as rice were limited and food ration cards were distributed. The land reforms in the 1980s which enabled farmers to grow and sell at their own responsibility and the vast support program for Vietnamese agriculture introduced in 1986 led to the huge growth of Vietnamese rice production. Nowadays, Vietnam is the second-largest rice-producing nation in the world.

In 2004, a new land reform was adopted which takes into account the changing economic conditions such as the fast development of industries and services in rural and peri-urban Vietnam. One challenge is the increasing difference in living conditions between the urban and rural population regarding income and the infrastructure provided.

By tying the progress of urban growth to the agricultural sector, a new rural area – with gradually modernizing socio-economic infrastructure and forms of production management coupled with the minimization of the existing and growing gap – will be achieved. In Decision No. 800/QD-TTg, the government stated its determination to reach a certain standard of eco-environment: rural areas with planned urban development and a democratic, stable rural community. To achieve these goals, several reforms, decreases and decisions have been introduced since 2008.

The Institute of Water Resources Planning estimated the annual water consumption of all sectors in the whole basin to be about 2.207 billion m^3. The agricultural sector is the largest water consumer (74%).

Water requirements are growing, with demand predicted to reach 2.553 billion m^3 by 2020. Although water use in agriculture has been reduced, this sector is still the largest water user.

2.7.2 Craft villages in the Red River Delta

For a long time, craft villages have been important for agricultural development in Vietnam. The first villages where the population at least seasonally pursued a craft existed back in the eleventh century, particularly in the area of Hanoi. A wide variety of goods were manufactured ranging from food and everyday products to skillfully designed silk and religious articles. Products were directly sold at local markets or made their way via retailers to the capital or even abroad. From their beginning, craft villages were closely intertwined with the economic network. In the communist era, they remained an important pillar of the Vietnamese economy. Since the introduction of the economic reform policy, the workforce and production volume of craft villages have continuously grown.

In the Red River Delta, handicraft villages were often found in certain separated areas or clusters that were linked to the capital via rivers and the extensive canal system. The production of goods often took place in the period between two rice crops, i.e. when farmers had time for another business. Thus, the development of craft villages was closely related to the cultivation of rice and the sophisticated irrigation system.

In the course of agricultural collectivization in the 1950s, private sector activities were restricted and craft villages were converted into cooperatives. However, rural production was hampered by the subsequent lack of raw materials. Sales were now under state control and the goods were mainly exported to Eastern European communist countries.

With the introduction of the economic reform policy in 1986, the production and distribution of goods was no longer state-controlled, and following the fall of the Berlin Wall, the markets in Eastern Europe were no longer available. The people in the craft villages faced the challenge of re-organizing their networks and, where appropriate, adjusting their production to modern market demands. New markets were made accessible and some craft villages changed their production completely, e.g. to recycled products.

Many of the craft villages are still organized in clusters that are economically intertwined. Often, specific production steps are allocated to individual villages, so that the villages cooperate rather than compete.

3 Materials and methods

3.1 Soil sample analyses

The two soil sampling campaigns were conducted in January 2012 and in June 2014. The first campaign was carried out to assess the total As concentration of the soil in the study area and the samples of the second campaign were treated by an SEF to estimate the mobile and bioavailable fraction.

In the first campaign, 13 samples from the paddy fields were taken from the upper 10 cm and from 20–30 cm depth (root zone). The samples were packed in airtight plastic bags and transported to Pirna/Germany. The pH of the soil samples was measured by a glass electrode in suspensions of soil and distilled water with a ratio of 1:5 (w/v).

For sample preparation, the soil samples were dried by oven until their weight was stable and unchanged at 105°C over 24 hours. For cooling, all the dried samples were placed in an exicator for 1 hour before weighing.

Gravimetric water content (w): The quantity of water contained in soil samples is defined by the ratio of the mass of water and the bulk mass.

$$\mathrm{w} = \frac{\text{mass of water}}{\text{bulk mass}} = \frac{(\text{bulk mass} - \text{mass of dried soil})}{\text{bulk mass}}$$

The range of w is from 0 to 1 (0 to 100%)

After drying, the soil samples were ground to powder with pestle and mortar and stored in plastic bags.

During the second campaign, 8 samples were taken from the root zone of the paddy fields. Sequential extraction fractionation was performed following the procedure described by (Wenzel et al. 2001). From each sample, 0.20–0.25 g of air-dried soil was placed in a 50 ml plastic centrifuge tube. The extraction steps are described in Table 85; each sample was treated twice. The separation of the liquid from the soil phase after each extraction step was performed by centrifugation at 5000 rpm for 15 minutes. The liquid was subsequently filtered with a filter paper. After filtration, the liquid was used to measure the concentrations of arsenic, iron and magnesium. The samples from step IV were centrifuged and then air-dried at 105°C for 24 hours. After the homogenization process, 0.2 g of each soil sample was used for microwave digestion in step V.

Table 5: Sequential extraction procedure

Steps	As fraction	Extractants	L/S	Extraction conditions
I	Nonspecifically sorbed	$(NH_4)_2SO_4$ 0.05M	1:25	4 hours shaking, 20°C
II	Specifically sorbed	$(NH_4)_2HPO_4$ 0.05M	1:25	16 hours shaking, 20°C
III	Amorphous and poorly crystalline hydrous oxides of Fe and Al	NH_4-oxalate buffer 0.2M	1:25	4 hours shaking, 20°C, in the dark
IV	Well-crystallized hydrous oxides of Fe and Al	NH_4-oxalate buffer 0.2M + ascorbic acid 0.1M, pH = 3.25	1:25	30 min in a water bath at 96°C
V	Residual phases	HNO_3/H_2O_2	1:50	Microwave digestion

The total As concentration in soil samples was determined by digesting 0.20–0.25 g of soil in a mixture of nitric acid and hydrogen peroxide. The samples were then digested in a MARS-5 microwave accelerated reaction system (CEM cooperation, USA).

After digesting, the samples were filtered through a 0.2 µm polytetrafluoroethylene (PTFE) membrane prior to analysis. All the microwave digestion experiments were performed in triplicate.

3.2 Well sampling

The water infrastructure study included the municipal water distribution network (tap water), private water supplies (drilled tubewells), rainwater used for domestic and production purposes, domestic sewage water, river water, and rainwater used for irrigation.

Over three years, seven water sampling campaigns were carried out: in December 2011, August 2012, April 2013, and four campaigns in 2014. The samples were taken from twenty randomly selected households, whereas the campaigns in 2014 were carried out on four wells. Fresh water samples were taken after having pumped up approximately 1 m^3 water. Temperature, pH, DO, EC and redox potential were measured on site following the Vietnamese Standards for fresh water analyses. Samples for TOC, heavy metals, cations and anions were collected following the Vietnamese Standards, stored in cooling boxes, and transported to the laboratory of the Vietnam Institute for Environmental Technology at the Vietnamese Academy for Science and Technology in Hanoi. For reasons of quality control, duplicate samples were measured.

3.3 Wastewater and sludge analyses

Wastewater samples were taken in April 2012, December 2012 and April 2013 during the 24 h measuring campaign by Hanover University (Meier 2015). COD_{total} and

$COD_{soluble}$ were analyzed by two measurements each hour and in a third measurement every three hours over a 24-hour measurement period. Some samples were determined directly in a temporarily established laboratory after they had been taken. Other parameters were determined at Bac Ninh Laboratory and in the laboratory of Hanoi University of Science.

Laboratory analyses were carried out using the following standard methods:

- COD: Hach Lange cuvette tests LCK 514 and APHA 5220
- NH_4-N: Hach Lange cuvette tests LCK 304 and LCK 303
- NO_3-N: Hach Lange cuvette tests LCK 339
- BOD_5: TCVN 6001:2008
- TS and VS: DIN 12880 and DIN 12879

The velocity of wastewater over 24 hours was measured by using a flowmeter (FP 101, Global Water Instrumentation Inc.) every 15 min. To determine low flows, the profile was reduced by constructing a wooden box similar to a venturi canal. The amount of wastewater was calculated by multiplying the velocity measured with the flowmeter by the volume of water. Parameters like temperature, pH, DO, TDS and salinity were determined using an HI 9828 instrument.

3.4 Food analyses

Rice, vegetables, fish, golden apple snails, eggs and meat are the only food which is grown and consumed in Dai Lam. The fresh samples were taken directly from the fields or gardens or were bought from farmers. All samples were stored in plastic bags and transported to the laboratory of the Institute of Waste Management and Contaminated Site in Pirna in Germany within two days. After being weighed, plant samples were separated into different parts: roots, leaves, branches and fruits. The roots were cleaned of soil; leaves and fruits were prepared as they are processed or cooked in Vietnam. From the animals, meat and liver was analyzed separately. Sludge and soil samples were not separated into fractions.

Preparation mainly followed the instructions in DIN 38414-S7. All samples were dried at 45°C for at least 24 hours. Afterwards, the samples were ground in a swinging mill (Retsch). 0.2–0.25 g of the sample was placed into Teflon vessels and 2 ml of H_2O_2 (30%) and 5 ml of HNO_3 (conc.) were added before the samples were digested in an MDS-2000 in steps up to 190°C and 10 bar. Subsequently, the samples were transferred with pure water into a 50 ml volumetric flask, and then filtered (0.2µm).

For ICP-MS measurement, the samples were diluted at least 1:10.

As standard, a Merck multi-element standard was used.

The samples were determined using an ICP-MS inductively coupled plasma mass spectrometer (Perkin Elmer-Sciex Elan DRC-e).

3.5 Site visit and field observations

Water and land use data collection was supported by site visits and field observations conducted in summer 2013. Information on agricultural areas, domestic areas, water bodies and water flow direction was gathered and documented.

3.6 Questionnaire

For this study, the main components of the hydrological water cycle in Dai Lam village were analyzed by directly collecting representative data sets including sampling campaigns and field surveys. In December 2011, a detailed survey was conducted in the village to enable a basic assessment of the socio-economic situation, including the characterization of the water-related infrastructure as well as of waste and water management practices. During the survey, six scientists interviewed 282 households representing 25% of the total households in the village. Additional interviews with the village leader, municipal leader, water workers, farmers, consultants and representatives of the irrigation company were carried out between March and July 2013.

4 Results

4.1 Soil samples

4.1.1 Total arsenic and total heavy metal concentrations

All soil samples were taken from the rice fields in the west of Dai Lam. The area appears to be very homogeneous and so the samples didn't differ very much from each other: the humic sandy loam is typical of the fluvial-deltaic sediments in this region. The color of the soils varied slightly from dark red brown to dark brown. The pH was mildly acidic.

The As concentration in the soil samples ranged from 3.67 to 33.21 mg/kg, the mean being 9.3 mg/kg (Figure 15), which is below the Vietnamese standard for agricultural soils (12 mg/kg). The concentration in the surface samples was lower than in the subsurface. The medium 50% of all samples had a concentration of between 6.6 and 9.5 mg/kg, which corresponds to the As concentration of other paddy fields (Phuong 2008).

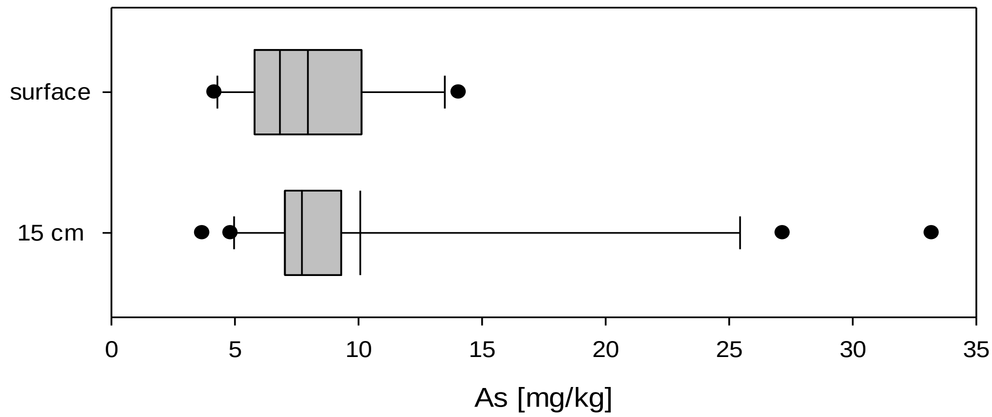

Figure 15: Arsenic concentrations in surface and root zone samples

The content of the organic matter in the soil is relatively homogeneous with 90% between 6.8 and 11.9% (Figure 16).

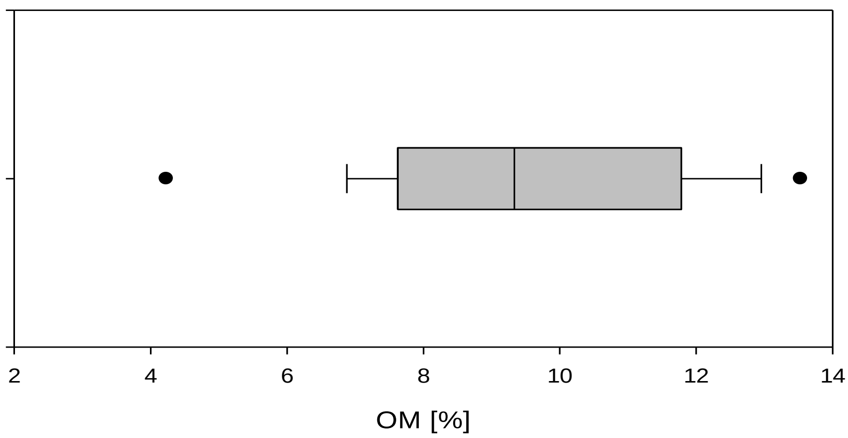

Figure 16: Block chart of organic matter in the soil samples

Fe and Mn are abundant in the soils of the Red River Delta and the soil samples from Dai Lam revealed high levels of both metals (Figure 16, Figure 17, Figure 18).

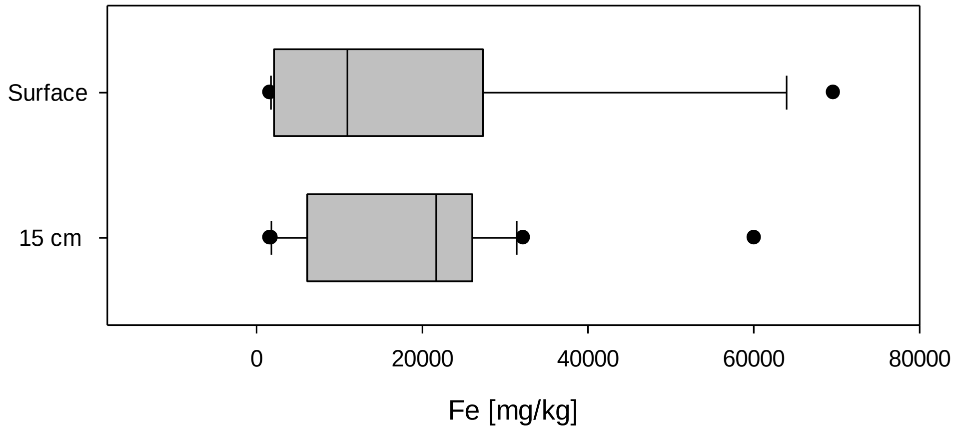

Figure 17: Block chart of the iron concentration in the soil samples

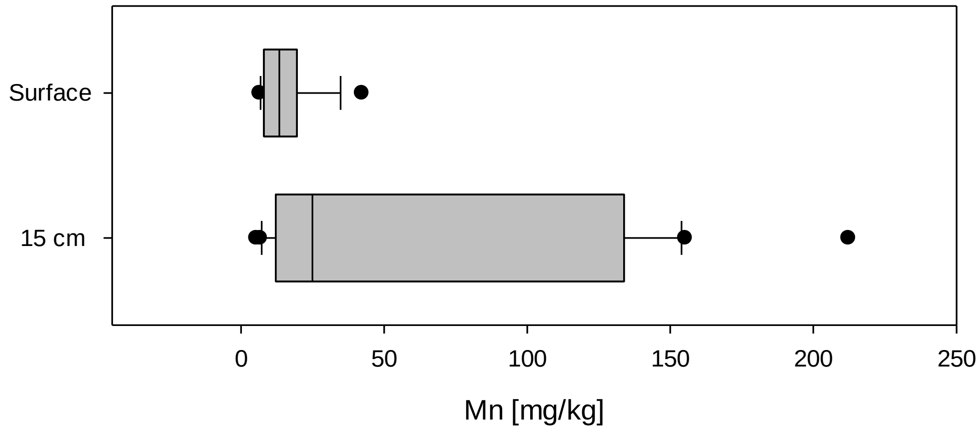

Figure 18: Block chart of the manganese concentration in the soil samples

Pearson's correlation of As, Fe, Mn, Cr, Co and Cd (annex) shows that whereas the upper soil layer doesn't reveal significant element correlation, the root zone samples show good correlation between all elements.

4.1.2 Sequential fractionation procedure

In order to assess the bioavailability of As in the study area, eight samples of the root zone were subjected to the sequential fractionation procedure after Wetzel. The five steps of the procedure reflect different orders of availability.

Sequential fractionation was carried out with eight randomly chosen paddy soil samples. The results of sequential fractionation are presented in Table 6. The As content of the nonspecifically sorbed fraction (F1) has very low levels of < d.l. to 0.04 mg/kg. The level of the specifically sorbed fraction (F2) is between <d.l. and 1.02 mg/kg. The amount which is assigned to the amorphous and poorly crystalline oxides of Fe and Al (F3) ranges between 1.97 and 3.0 mg/kg and the amount of the well-crystallized oxides

of Fe and Al (F4) between 1.17 and 12.99 mg/kg. The residual phase (F5) has As levels between 1.22 and 3.56 mg/kg.

Table 6: Results of sequential fractionation

Soil	***As***				
	mg/kg				
	F1	*F2*	*F3*	*F4*	*F5*
DL 002	0*	0.48	1.97	1.96	1.37
DL 003	0.04	0.62	2.23	1.49	2.74
DL 004	0.01	0.71	2.87	2.87	1.22
DL 007	0.01	0.74	2.39	1.17	1.47
DL 008	0	1.02	2.92	12.99	3.56
DL 011	0*	0.69	2.01	3.17	2.2
DL 012	0*	0*	3	3.37	2.07
DL 014	0*	0.249	2.04	2.99	1.73
Min	0	0	1.97	1.17	1.22
Max	0.04	1.02	3	12.99	3.56
Mean	0.01	0.56	2.43	3.75	2.04

Figure 19 shows that the fractions in relation to the total As content appear to be relatively homogeneous. The two fractions which could represent the bioavailable fraction (F1 and F2) are low (10%), the residual fraction (F5) accounts for 25% of the total As, and so the proportion of As which is associated with the crystalline and amorphous iron hydroxides (F3 and F4) accounts for 65%.

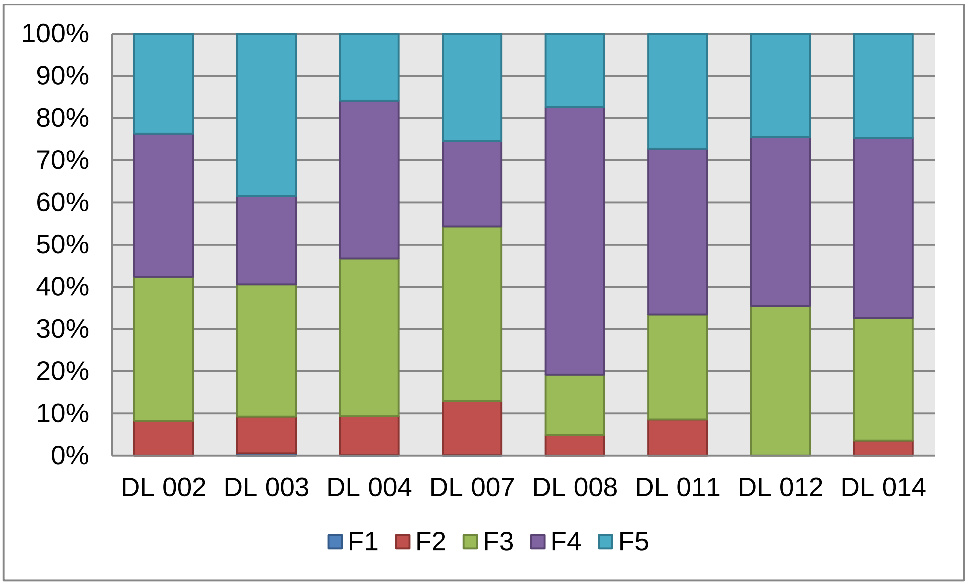

Figure 19: Partitioning of the As content in eight paddy soil samples

4.2 Arsenic in the water cycle in Dai Lam

4.2.1 Groundwater analyses

The tube wells were constructed in 2008 in order to protect the population from the unsafe water supply. Most wells pump the water from a depth of 17–22 m, which originates in the lower Holocene aquifer. The water from the tube wells isn't considered safe by most interviewees due to its yellowish color and 'fishy' smell, and many households avoid using the tube well water for cooking and drinking.

The main sampling campaigns were carried out in winter 2011 (during the dry season), in summer 2012 (during the wet season), and in spring 2013 (at the end of the dry season). In 2014, additional sample campaigns were carried out in order to study the temporal variation of the arsenic concentration. The statistical key data of the arsenic concentrations are presented in Table 7, the variation of the concentration is shown in Figure 20 and the spatial distribution of the sampling points is shown in Figure 21. The three main campaigns showed varying arsenic concentrations. In winter 2011, the mean concentration slightly exceeded the international threshold value of 10 µg/l and the highest concentration was 28 µg/l. In summer 2012, the mean value was 44 µg/l and the maximum concentration 106 µg/l, whereas in spring the mean arsenic concentration was 17 µg/l. Adjacent to these obviously high amplitudes, a remarkable difference in the variation of the values was observed. The standard deviation of the three sampling campaigns showed that the sampling campaign in summer 2012 had the highest variance. Five additional sampling campaigns of four tube wells in 2014 exhibited increased arsenic concentrations.

Table 7: Statistical key data for arsenic [µg/l] in well samples

	2011	2012	2013	04/2014	06/2014	08/2014	10/2014
Mean	10.5	44	17.1	8.75	7	6	7.75
Median	8	45	8	8.5	5.5	4.5	
Std. deviation	8.56	33.70	16.50	3.86	5.94	4.24	4.64
Min	1	6	1	5	2	3	3
Max	28	106	52	13	15	12	14
Number of samples	20	24	20	4	4	4	4

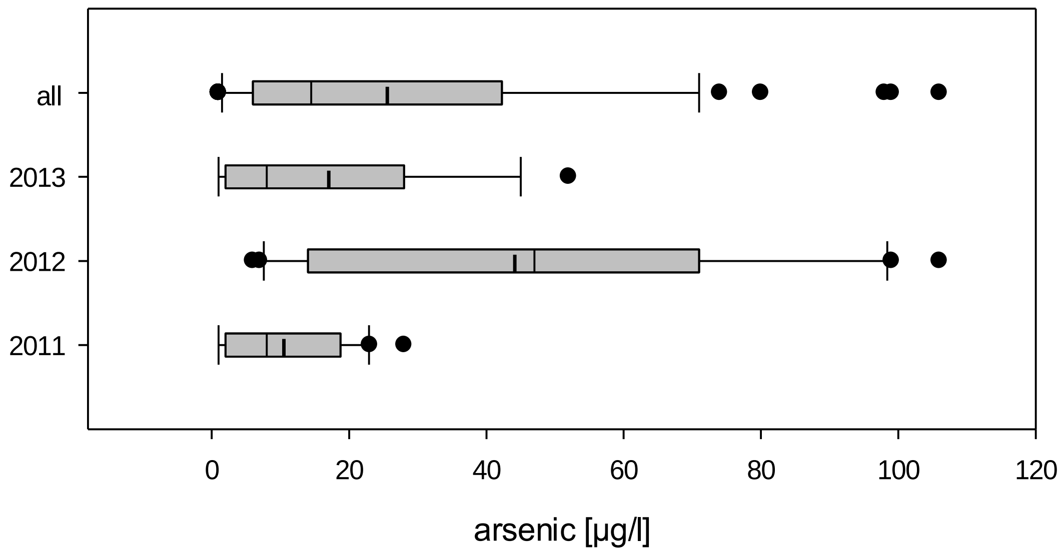

Figure 20: Block box diagram of the As concentrations in the wells

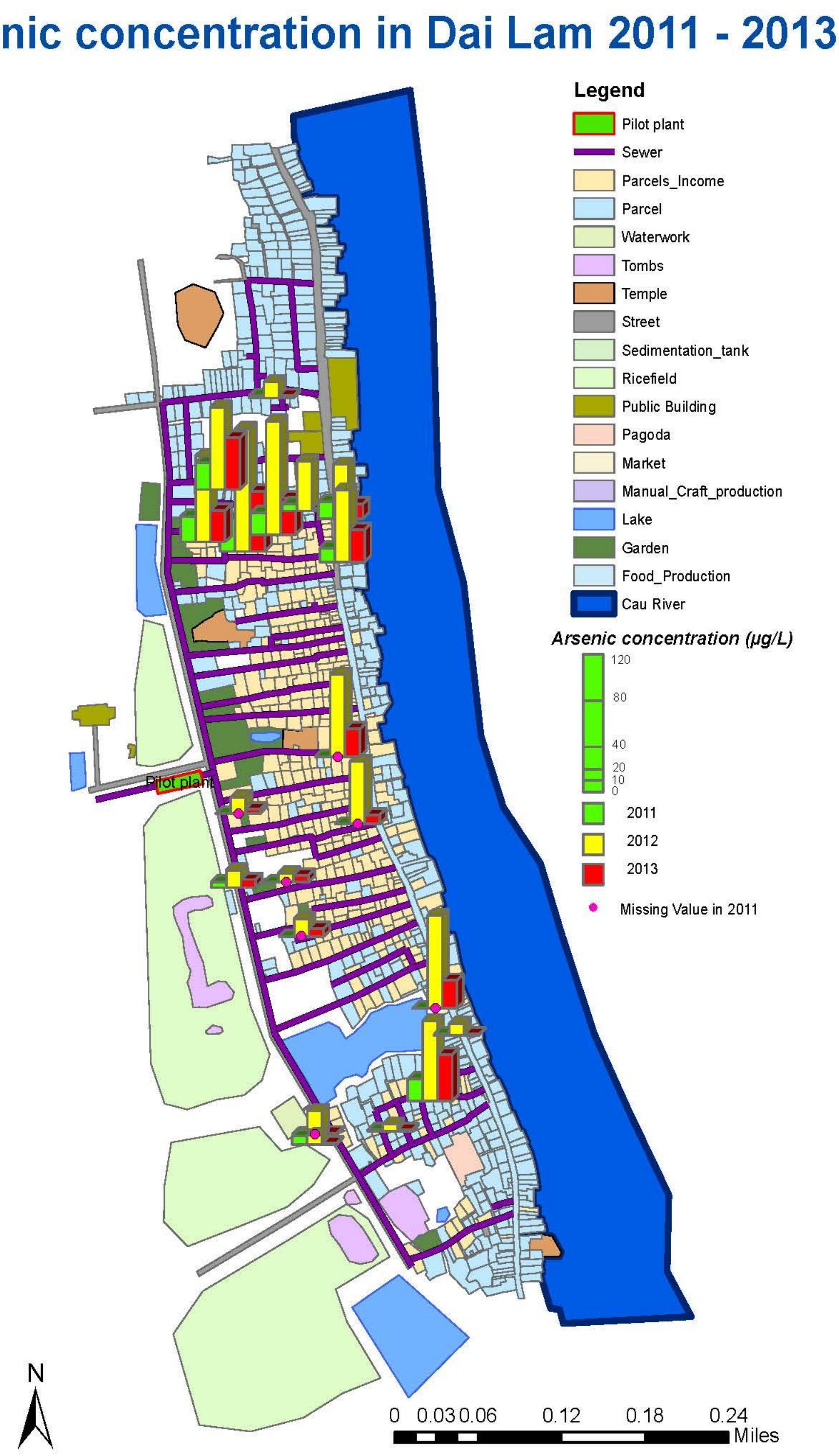

Figure 21: Arsenic concentration in well water in Dai Lam

Apart from the arsenic concentration, other parameters were analyzed such as NH_4^+, ORP, EC DO, TOC, NO_3^-, NO_2^-, SO_4^{2-}, PO_4^{3-}, Fe, Mn, Pb and coliforms. The mean values and the standard deviation of the different parameters are shown in Table 8. The characteristics of the water are summarized as follows:

- The lowest water temperature was measured in April 2013 at 23.8°C and the highest was measured in summer 2012. The difference of 2°C correlates with the seasonal cycle of shallow aquifers in the Red River Delta (Giang et al., 2014), which shows a peak in June–September.
- The mean pH value of the three campaigns also varies slightly but significantly from 6.8 in 2011 to 7.1 in 2013. However, the values are in the expected range.
- The dissolved oxygen is quite low, which indicates oxygen-consuming processes in the upper aquifer.
- The electrical conductivity was remarkably high in all three campaigns. The samples from Dai Lam indicate that the groundwater has been affected by anthropogenic activities.
- The ORP values highlight the reducing conditions in the aquifer.
- The sulfate values meet international standards.
- The ammonia exceeds the Vietnamese standard for drinking water of 0.1 mg/l. The high values indicate that the hygienic conditions are risky and that the aquifer is affected by intensive agricultural or industrial activities.
- The TOC results indicate an increased mean value in 2011, underlining the higher pollution level of the aquifer. In 2012 and 2013, only 20% of the wells showed a TOC value higher than 10 mg/l.
- All three campaigns reveal very high iron concentrations. The results exceed the international threshold values of 0.2 mg/l (EU) and also the Vietnamese threshold of 5 mg/l. Enhanced iron concentrations are common in aquifers with reducing conditions and low oxygen concentrations.
- Manganese also exceeds the international standards in all three campaigns (WHO: 0.4 mg/l, Germany: 0.05 mg/l, Vietnam: 0.05 mg/l). Like the iron concentrations, manganese concentrations are often elevated under reducing groundwater conditions.

Table 8: Mean values and standard deviation of well samples

Year		2011	*SD*	2012	*SD*	2013	*SD*
T	[°C]	25.5	*0.3*	25.9	*0.5*	23.8	*1.0*
pH		6.8	*0.1*	7.1	*0.2*	6.9	*0.3*
DO	[mg/l]	1.17	*0.79*	1.28	*1.49*	1.77	*0.77*
EC	[µS/cm]	785	*324*	765	*332*	633	*180*
ORP	[mV]	–139	*31*	–149	*32*	–127	*36*
SO_4^{2-}	[mg/l]	41.3	*36.2*	31.8	*23.0*	28.1	*41.9*
NH_4^+	[mg/l]	5.4	*1.8*	6.6	*3.4*	5.0	*3.6*
TOC	[mg/l]	14.9	*5.0*	7.2	*4.6*	7.4	*3.6*
As	[µg/l]	10.5	*8.6*	44.2	*32.3*	17.2	*16.5*
Fe	[mg/l]	4.84	*3.20*	32.77	*20.89*	16.48	*11.67*
Mn	[mg/l]	0.15	*0.12*	1.01	*0.46*	0.45	*0.42*
coliforms	MPN/ 100mL	4		5.5		10.05	

4.2.2 Water use in Dai Lam

4.2.2.1 Water supply

Drinking water for the inhabitants of Dai Lam is supplied by an existing waterworks. Most households are connected to the pipe network and have access to clean water. The waterworks largely meets the national standard (TCXDVN 33/2006). It consists of only one tank with a volume of 180 m³. According to the household survey in Dai Lam (2012), 89% of the households use tap water, especially for cooking and drinking. According to the national set of criteria on new rural development in the Red River Delta (criteria 17.1, Decision No. 491/QD-TTg), 90% of the people are estimated to have access to hygienic water (‘Quality suits national standards’, Decision No. 09/2005/QD-BYT). Although the water is treated in the waterworks, the majority of households (at least 78%) re-filter the tap water. Many people feel safer using a filter due to the instable water quality: the water frequently smells strange or has a yellow tinge. Big tanks filled with fine yellow sand mixed with charcoal are the most convenient filters. These filter tanks also serve as storage tanks in order to insulate homes from interruptions to the water supply (Nunweiler 2012).

The water is pumped up from 45 m. After treatment in the waterworks, it is pumped into the households. The water supply system consists of main pipes (90 cm in diameter, 4 km in length) which are connected to smaller pipe branches leading into the lanes (60 cm in diameter, 7 km in length). The water then is distributed among the households (48 cm, 27 cm and 21 cm diameter pipes). The pipe network is not maintained frequently, as a result of which leaking pipes and low water pressure are common problems. The total output of drinking water reaches 600 m³/day (CTIC 2011).

Last year, the organization of the waterworks changed. It is no longer financed by the village (centralized fees). Instead, the employees in the waterworks started to collect user fees by themselves. According to an interview with waterworks employees, this new system works out well and most people pay reliably.

In the master plan, the values for the Tam Da municipality as a whole are estimated as follows:

Table 9: Water consumption of total Tam Da Municipality, master plan, 2012

Consumer	Supply standard		Unit	Total demand (m^3/24h)	
	2015	2020		2015	2020
Living activities (LA)	100	120	l/person/24h	1310	1806
Public use	20	30	% LA	262	542
Fire (1 fire/2h)	15	15	l/s	324	324
Tree watering, road cleaning	10	12	% LA	131	217
Total				**2,027**	**2,889**
Reserve, leakages	10	20	% of total	203	578

These assumptions allow the values for Dai Lam village to be estimated.

In the master plan, the standard value of water use for living activities is defined as 100 l/person/day in 2015 (Table 9). This value is borne out by the results of the household survey in 2012. The total water demand in the municipality amounts to 470 m^3/d on average. Until 2020, water demand for living activities is predicted to constantly rise to 120 l/person/day. Moreover, at least 1,420 new inhabitants are expected to move to Dai Lam by 2020 (not counting births), increasing total water consumption to at least 805 m^3/d. Furthermore, the demand for water supply in public and cultural buildings and for public activities is set to rise while the development of new public amenities, as announced in the master plan, will exacerbate this increase.

The analysis of the interviews in 282 households in Dai Lam carried out in December 2011 showed that the water currently used for domestic and production purposes originates mainly from tube wells and the communal distribution network (78.4%), although several households also use water from rainwater tanks (figure 22). In addition, more than 10.9% of the households use only the tube well water for all domestic purposes like drinking, cooking, washing and flushing.

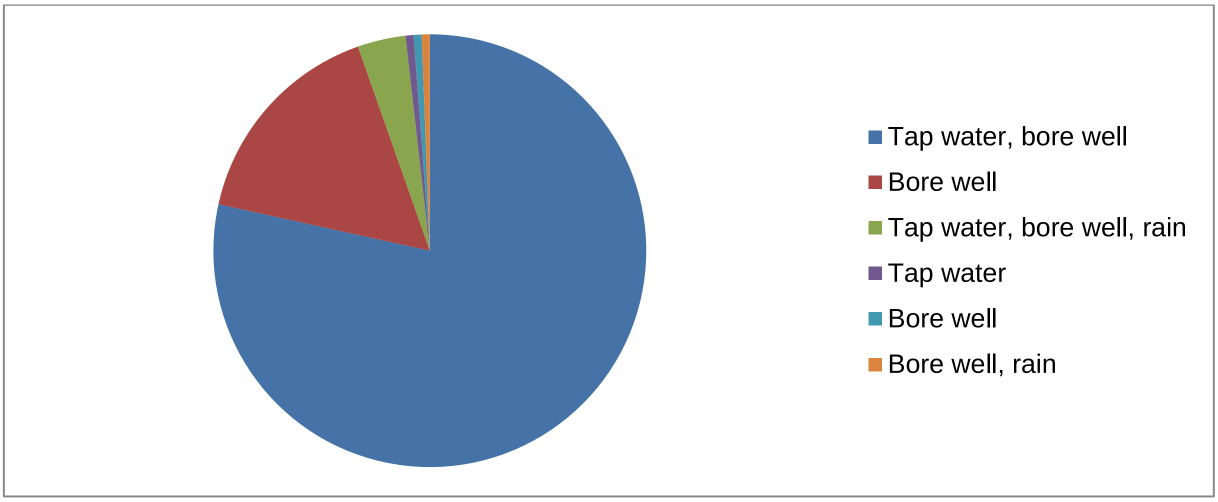

Figure 22: Sources of domestic water by number of households (%)

Water utilization in households

Figure 23 shows the different purposes of water usage in domestic households. Piped water is used mainly for drinking and cooking, while tube well water is employed in most households as service water. However, more than 25% of the households still use tube well water for all domestic purposes, including drinking and cooking. Eighty percent of these households use sand or coal filters, while the remaining households don't practice any pre-treatment. Particular attention needs to be paid to the use of tubewell water as production water. This applies to almost 19% of the households, which use tube well water in noodle and wine production as well as pig and poultry farming. The type of water used by households and small enterprises hinges on the associated costs. Then again, consumers are aware of the risks of tube well water and prefer to use piped water for drinking and cooking. This must be taken into account when the water supply system is modernized and the wells are sealed because in rural areas the profit margins are still very small and piped water could be significantly more expensive for water-consuming enterprises (Noel 2010).

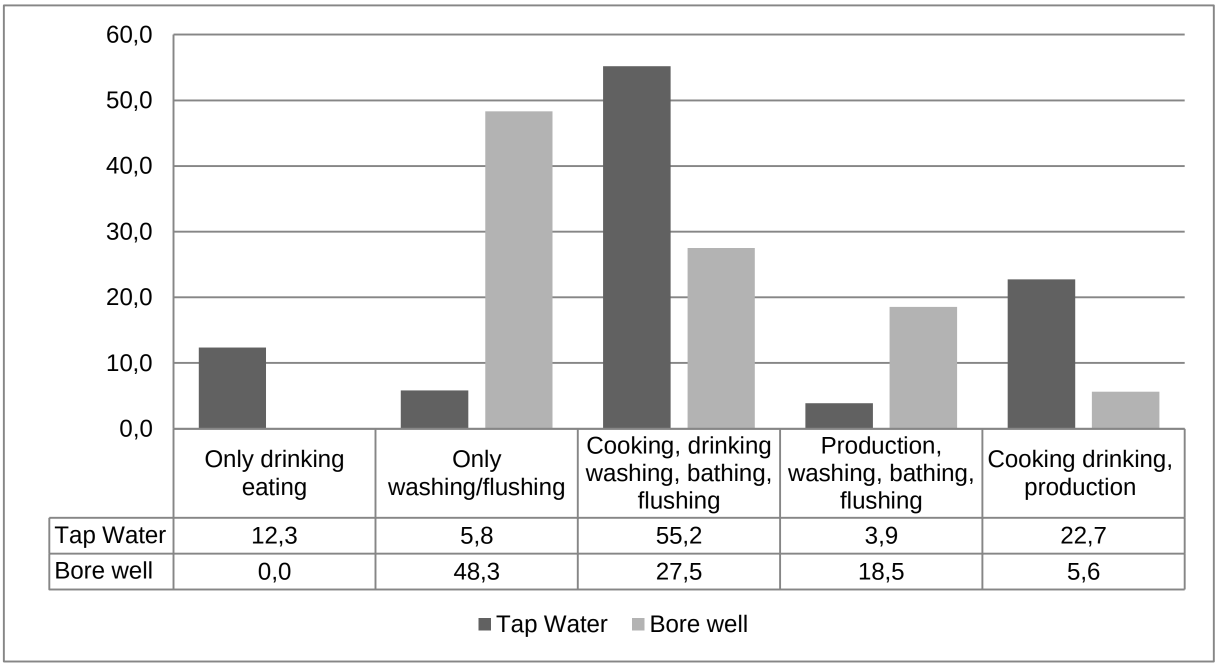

Figure 23: Water utilization in domestic households (%)

While tap water consumption can easily be assessed thanks to the water meters installed, the tube well water had to be calculated from the pumping times throughout the day. In total, 1,238 m³ of water is pumped and withdrawn daily from the public water supply, yielding a water consumption of 255 l/person. Figure 24 depicts the mean daily water consumption in the village: about 34% originates from the waterworks, 66% is pumped up by the bore wells.

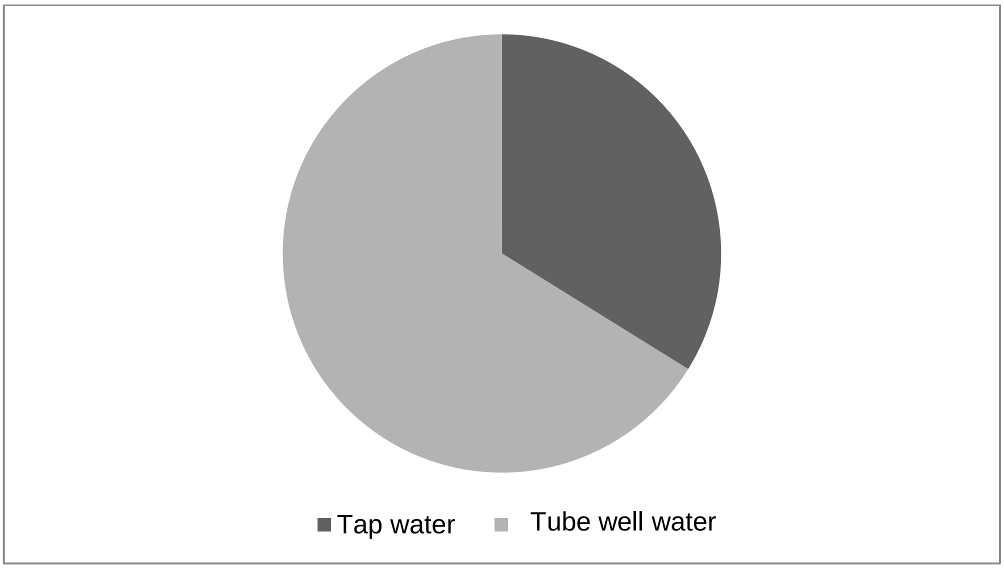

Figure 24: Estimated daily water consumption

4.2.3 Wastewater in Dai Lam

Three wastewater sampling campaigns were carried out in Dai Lam in 2012 and 2013 by the University of Hanover to investigate the amount and quality of the wastewater.

The results of the raw wastewater analyses shown below in Table 10 are part of detailed investigations in the scientific work by Sebastian Meyer (2015). The investigations show a range of Q_{total} of 73.3 and 140.8 l/person/d. The chemical oxygen demand (COD) of the soluble phase varies between 347 mg/l and 487 mg/l.

Table 10: Results of the wastewater analysis (Meyer, 2015)

		Apr 12	Dec 12	Apr 13
Q_{total}	m^3/d	669	445	348
$Q_{soluble}$	$m^3/d/cap$	140.8	93.7	73.3
COD_{total}	mg/L	617	958	845
$COD_{soluble}$	mg/L	400	347	487
NH_4-N	mg/L	188	-	143
NO_3^-	mg/L	0.96	0.67	0.64
pH		7.4	7.6	7.2
TDS	mg/L	1297	1162	372
Conductivity	mV	2598	2239	765

Between December 2011 and January 2013, four samples were taken from the central wastewater inlet into the canal. The As concentrations are shown in Table 11. The heavy metal concentration and COD content in the soluble phase are contained in Annex 9.2.4.

Table 11: As analyses of wastewater

		Dez-11	Apr-12	Oct-12	Jan-13
As	[µg/L]	7.9	12.5	10.3	15.6

4.3 Arsenic in sewage sludge

During the INHAND project, a three-step waste and wastewater treatment plant was installed in Dai Lam in 2012 by VIS GmbH and Herbst Umwelttechnik GmbH. The aim was to implement an integrated waste and wastewater treatment concept. The plant consisted of an aerobic SBR unit, an anaerobic reactor to produce biogas from the sewage sludge and organic waste from the village, and a digestate drying facility. The functional parts are shown in Figure 25. The sludge products are marked C (sewage sludge), F (direct outlet of the anaerobic reactor), G (digestate) and H (dried digestate).

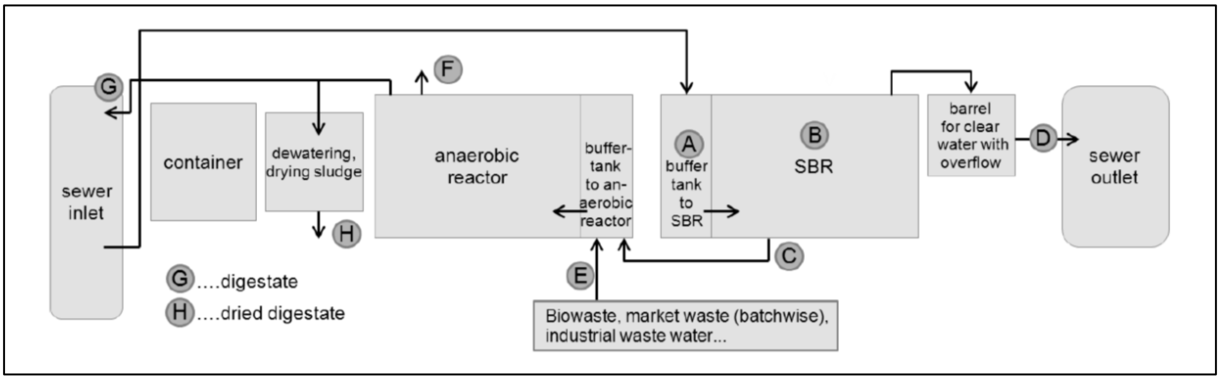

Figure 25: Diagram of the three-step pilot plant (Meier, 2015)

Table 12 shows the average results of the sludge samples of the pilot plant. The average of all samples is 30 mg/kg and the variation of the results is low.

Table 12: As content in the flow streams of the pilot plant

Sample	A	B	C	D	F	G	H
mg/kg and µg/l (liquid)	11.33 (l)	24.75 (l)	31.76	3.34 (l)	27.55	28.80	30.53
SD	0.62	0.52	1.83	0.14	1.75	1.04	1.14

4.4 Arsenic in manure samples

On average, each household breeds 10 pigs per year in the village of Dai Lam – making in total 10,000 pigs annually. The piggery was originally a by-product of the wineries: the soaked rice grains (mash) are a nourishing food for pigs and so almost every craft household bred some pigs. Recently, the production of rice wine has become less lucrative than the piggeries, which is why many pigs are fed with additional food and only some pigs are given the soaked rich grains as their principal food.

In this study, two manure samples were taken and analyzed (rice from the area of Dai Lam.

Table 13) (heavy metal results in Annex 9.2.5). Pig 1 was fed with industrial food, while Pig 2 was fed with soaked rice from the area of Dai Lam.

Table 13: As concentration in manure samples

	Pig 1	Pig 2
As [mg/kg]	0.32	1.25

4.5 Arsenic in food samples

4.5.1 Rice

The rice grain samples were taken from six different sites around the village of Dai Lam. In four samples, the concentration was below the detection limit. The maximum value was 0.59 mg/kg and the mean value was 0.23 mg/kg. The maximum value in the leaves was 8.20 mg/kg and the mean was 6.56 mg/kg. The stems had a lower As concentration (maximum 6.1 mg/kg and a mean concentration of 3.21 mg/kg). Samples with mixed stems and leaf compartments showed a mean value of 2.3 mg/kg.

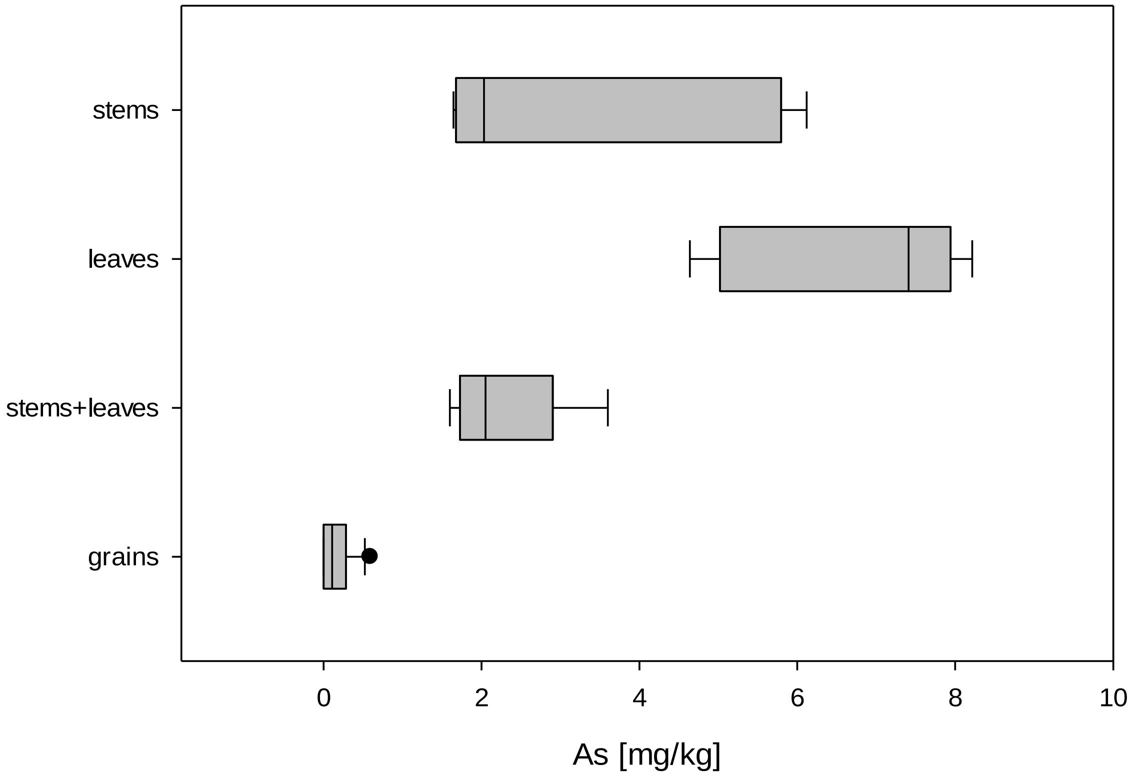

Figure 26: arsenic concentration in rice plants (stems, leaves and grains)

Table 14: arsenic concentration in rice plants (stems, leaves and grains)

	Mean [mg/kg] dw	SD [mg/kg]
Rice grains (n=11)	0.23	0.18
Leaves (n=9)	6.56	1.53
Stems (n=9)	3.21	2.03
Mixed leaves and stems (n=8)	2.30	0.70

In Dai Lam, seven out of nine samples exceeded this value.

4.5.2 Arsenic in leaf vegetables

One of the most commonly grown vegetables in Dai Lam is *ipomoea reptans*, also known as water spinach, leafy vegetable kalmi or morning glory. But also other feafy vegetables are grown in the gardens for own consumption (Figure 27).

Figure 27: Bed of water spinach in Dai Lam, white cabbage, water spinach, kohlrabi, lettuce

Three mixed samples from three beds of water spinach were taken and the edible parts (stems and leaves) were dried and their heavy metal concentrations analyzed. The water content of the stems and leaves was 91.5%. In order to make a reliable risk assessment, one part of the samples was washed before the preparation of samples and the other part was left unwashed.

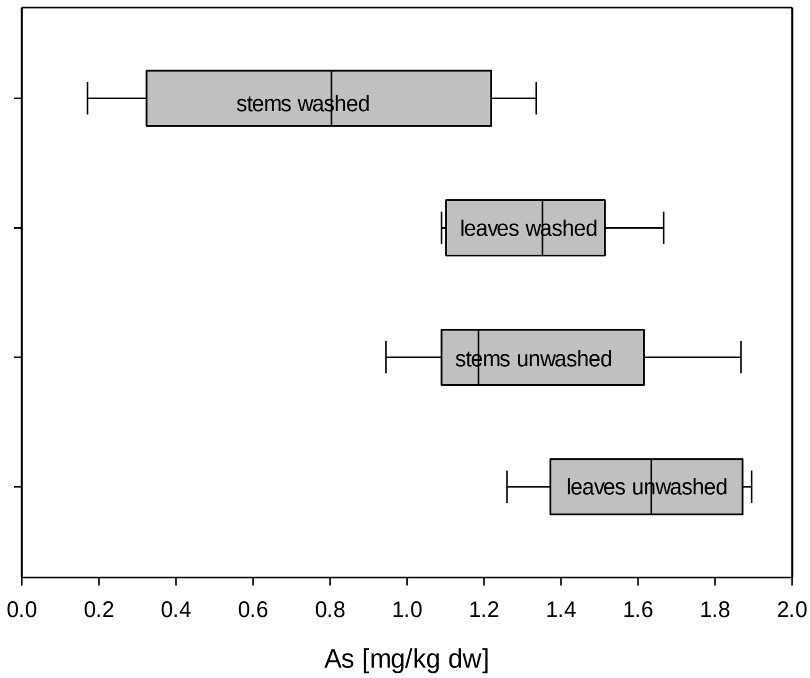

Figure 28: Variation of As in water spinach samples (n=9)

Figure 28 shows that the unwashed samples have higher As concentrations than the washed samples. The mean As concentration in the unwashed samples was 1.47 mg/kg, 43% higher than in the washed samples (1.03 mg/kg). The mean difference between the washed and unwashed samples (0.6 mg/kg) of the stems was more significant than the leafy part of the plants, where the difference was 0.29 mg/kg. Additionally, the leaves showed a higher As concentration than the stems.

Table 15: Pearson's correlation analysis of As with other heavy metals in water spinach (C: correlation coefficient, p: Pearson's value)

		Al	Fe	Mn	Co	Cu	Cd
stems washed	C	-0.701	-0.354	0.996	-0.849	0.718	0.697
	p	0.0355	0.35	1.04E-08	0.00375	0.0293	0.0368
leaves washed	C	-0.924	-0.928	0.986	0.83	-0.89	-0.895
	p	3.65E-04	3.05E-04	1.12E-06	5.60E-03	1.32E-03	1.11E-03
stems unwashed	C	0.458	0.435	0.049	0.299	0.432	0.824
	p	0.215	0.242	0.9	0.434	0.245	0.0063
leaves unwashed	C	0.948	0.95	-0.712	-0.784	0.559	0.402
	p	9.92E-05	8.72E-05	0.0313	0.0125	0.118	0.284

The Pearson's correlation factors of As with several heavy metals are presented in Table 15. The results reveal only a significant correlation between As and Mn and Al for the washed samples. The other coefficients don't prove a correlation: for cobalt, copper and cadmium the Pearson's value for the washed samples is lower than 0.05, although the algebraic sign is different.

Samples of white cabbage (*Brassica oleracea var. capitata f. alba*) showed As concentrations between below the detection limit and 3.54 mg/kg (dw). Lettuce exhibited a concentration of 2.04 mg/kg (dw) and kohlrabi samples had a concentration between below the detection limit and 0.26 mg/kg (dw) (Table 16).

Table 16: As concentrations in vegetable in dried and wet samples from Dai Lam

	dw [mg/kg]	ww [mg/kg]
Kohlrabi	<0.002–0.026	<0.002–0.0025
Lettuce	1.92–2.28	0.13–0.16
White cabbage	<0.002–3.3	<0.002–0.49
Water spinach	0.34–1.64	0.03–0.14

4.5.3 Arsenic in poultry products

Many households in Dai Lam raise chicken for their own consumption. In the survey, 88 out of 280 households stated that they raised chicken for their own consumption of the eggs and meat. The chickens mostly lived in the courtyard and were fed with leftovers from meals of the inhabitants or pigs. In this study, the flesh and the liver of three chickens were analyzed regarding their As and heavy metal content (Table 18).

Chicken 1 was fed with the family's everyday waste. It lived on braised rice and wheat grains and cooked meals and it drank filtered well water. The weight of the chicken was 1.3 kg and the age was 5 months.

Chicken 2 was fed with waste from the piggery. The age of the chicken was 5 months old and looked rather underfed, weighing just 0.95 kg. It didn't lay any eggs.

Chicken 3 lived outside the courtyard and survived in the grassland on insects, worms, grains and waste. The chicken weighed 1.4 kg and was 4 months old.

Table 17: As in poultry products [mg/kg] dry weight and wet weight

dw	Meat	Liver	Eggshell	Egg white	Yolk
Chicken 1	ND	0.376	ND	0.256	0.028
Chicken 2	ND	0.496	ND	0.249	1.428
Chicken 3	ND	0.987	ND	0.112	0.134
ww	**Meat**	**Liver**	**Eggshell**	**Egg white**	**Yolk**
Chicken 1	ND	0.135	ND	0.003	0.013
Chicken 2	ND	0.133	ND	0.028	0.675
Chicken 3	ND	0.261	ND	0.015	0.063

4.5.4 Arsenic in pork samples

In this study, one pork meat sample and three liver samples were analyzed. The pigs were all raised in Dai Lam. Pig 1 was four months old and was fed with soaked rice grains and kitchen waste; the age of Pig 2 is not known, but it was fed with industrial processed food; Pig 3 was 3.5 months old and fed mainly with soaked rice. The results are presented in Table 18. Note the considerable differences between them.

The high As concentration in liver 3 is proved by the analysis results for heavy metals (Annex).

Table 18: As in pork liver and meat [mg/kg] ww

Liver	Pig 1	Pig 2	Pig 3	Pork meat
As	0.42	0.01	0.79	0.07

4.5.5 Arsenic in snails

During the study run time, the high occurrence of golden apple snails was observed. The habitat is the paddies (Figure 29) as well as ponds and other wet beds (water spinach). Ten samples were taken from randomly chosen paddies near the village. Besides the As content, Fe, Mn, Cr, Co and Ni were also determined (table 19, Figure 30). All metals were detected in high concentrations. The As content in the snail samples differed greatly from shell to core and in 8 out of 10 samples the core of the snail had a higher As concentration than the shell.

Figure 29: Golden apple snails in the paddies

Table 19: Statistical metal data of snail samples [mg/kg ww]

	As		Fe		Mn		Cd		Cr		Ni	
	Shell	Core	Shell	Core	Shell	Core	Shell	Core	Shell	Core	Shell	Core
Mean	0.37	1.01	966	1966	256.6	331.7	0.92	0.29	14.1	1.2	105.3	16.4
SD	0.36	0.45	308	792	115.6	171.4	0.29	0.18	5.2	0.6	26.0	20.5
Max.	1.14	1.74	966	1966	409.2	640.6	1.35	0.71	26.4	2.5	150.3	64.7

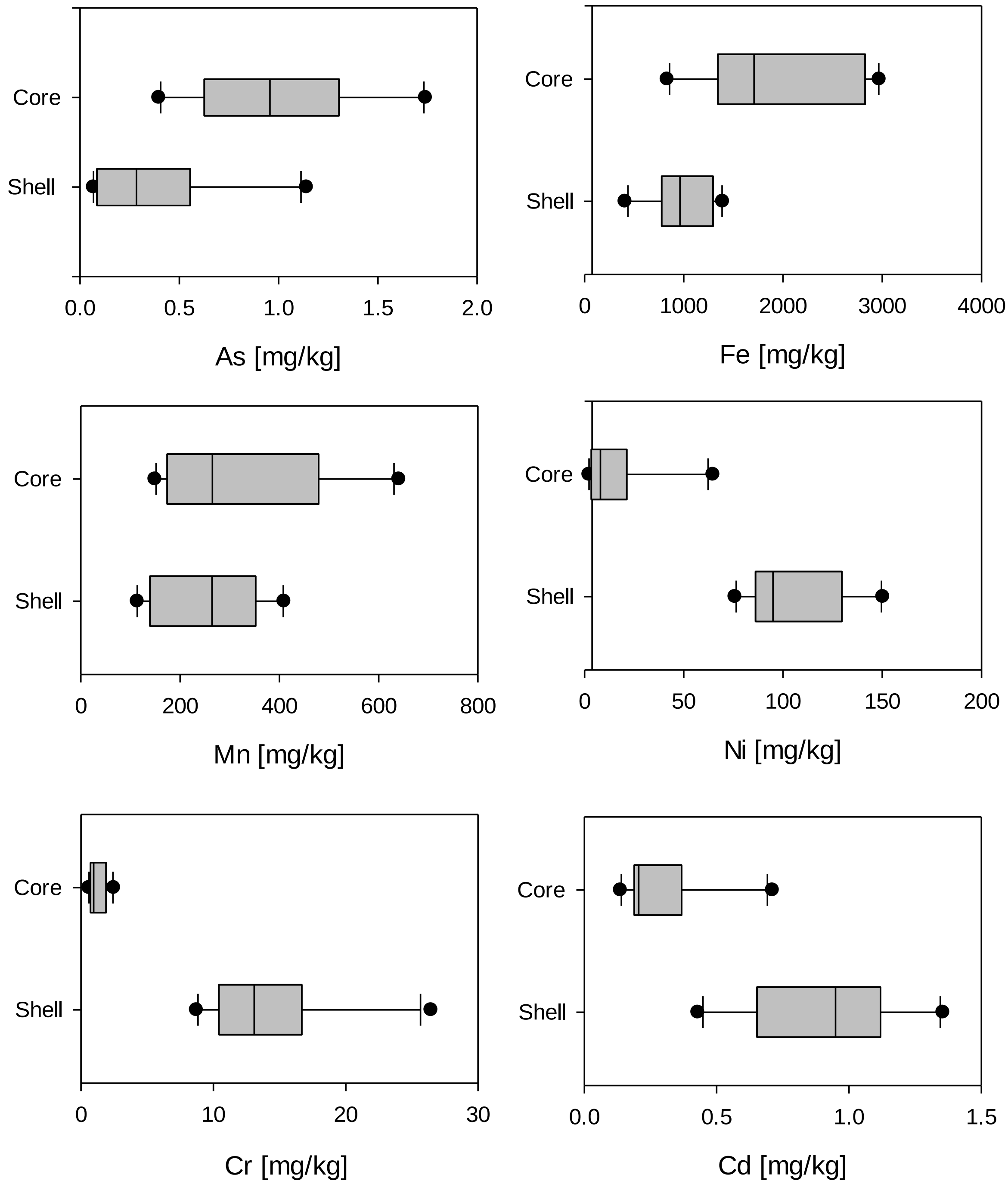

Figure 30: As and metal concentrations in the shell and core of GAS [mg/kg ww]

This characteristic can also be observed for Mn and Fe, whereas Ni, Cd and Cr accumulate in the shell.

4.6 Economic and demographic development potential

Dai Lam is one of the four hamlets in the province of Tam Da and has about 5,300 inhabitants in 1,240 households (4.27 persons per household). Dai Lam faces high population growth. From late 2011 until late 2012, its population grew by 2.48% (Table 20). Dai Lam's is undergoing above-average population growth, which in the country as a whole is 1.054% (CIA, 2013). In fact the population of Dai Lam has undergone mean annual growth of 2.56% since 2009.

Table 20: Population of Dai Lam per year and year-on-year growth in %, (Tam Da, Population Report 2012)

Year	Total	Female	Male	House-holds	Increase to previous year in %
2009	4,905	2,482	2,423	1,170	-
2010	5,027	2,567	2,460	1,197	2.49
2011	5,164	2,459	2,705	1,196	2.73
2012	5,292	2,607	2,685	1,237	2.48
Mean					2.56

The increase in population can be attributed to both a high birthrate and the high migration rate. The Tam Da municipal leadership explains this development to result from the effects of the new industrial area located close to Dai Lam in Yen Phong district. The leadership is convinced that the Yen Phong 1 Industrial Zone (IZ) will promote industrialization and rising incomes in the villages. According to the master plan, the population growth of the Tam Da municipality until 2020 is estimated as follows:

Table 21: Forecast of the total population of Tam Da Municipality (master plan, 2011)

Status	Year	Inhabitants	Increase
Current	2011	11,737	
Forecast	2015	13,109	1.372
	2020	15,049	3.312
Exponential growth rate: 1.3%			
Logistic growth rate: 1.5%			

The village of Dai Lam is the most populated village in the Tam Da municipality. About 45% of all the inhabitants of the Tam Da municipality live in Dai Lam village. Dai Lam is the village in the municipality that is predicted to undergo the largest growth and the largest planned residential area. Judging by the current distribution, Dai Lam will have 5,900 people in 2015 and about 6,800 inhabitants in 2020. However, the specific distribution of Tam Da's population among the four different villages needs to be noted.

The household inventory at the end of 2011 showed that the average income in Dai Lam was 4.8 million/household VND (€177) and that 41.5% of the households had a monthly income between 1 and 5 million VND (Figure 31).

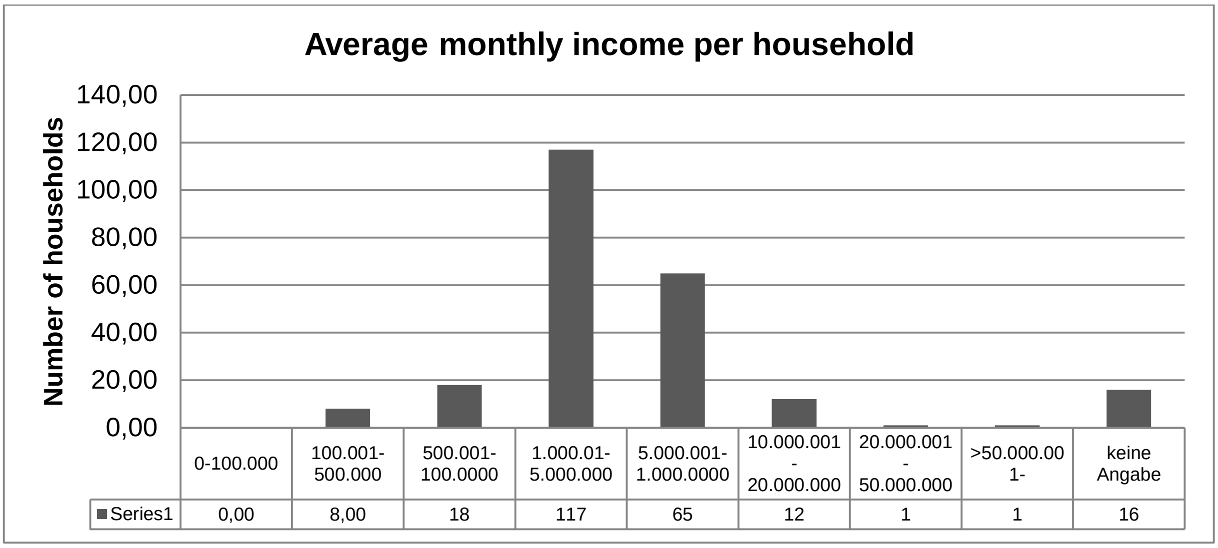

Figure 31: Monthly income per household

The main income source is agriculture; 122 of 280 (44%) households practice arable farming, although only 15% of the respondents make their living solely from farming. Eighty-three percent of the respondents practice at least one additional occupation: 27% have their own small business in Dai Lam or in the next city. These businesses are food processing or small shops. One third of the villagers have other occupations in the municipality, in construction companies, or in one of the new industrial zones (Figure 32).

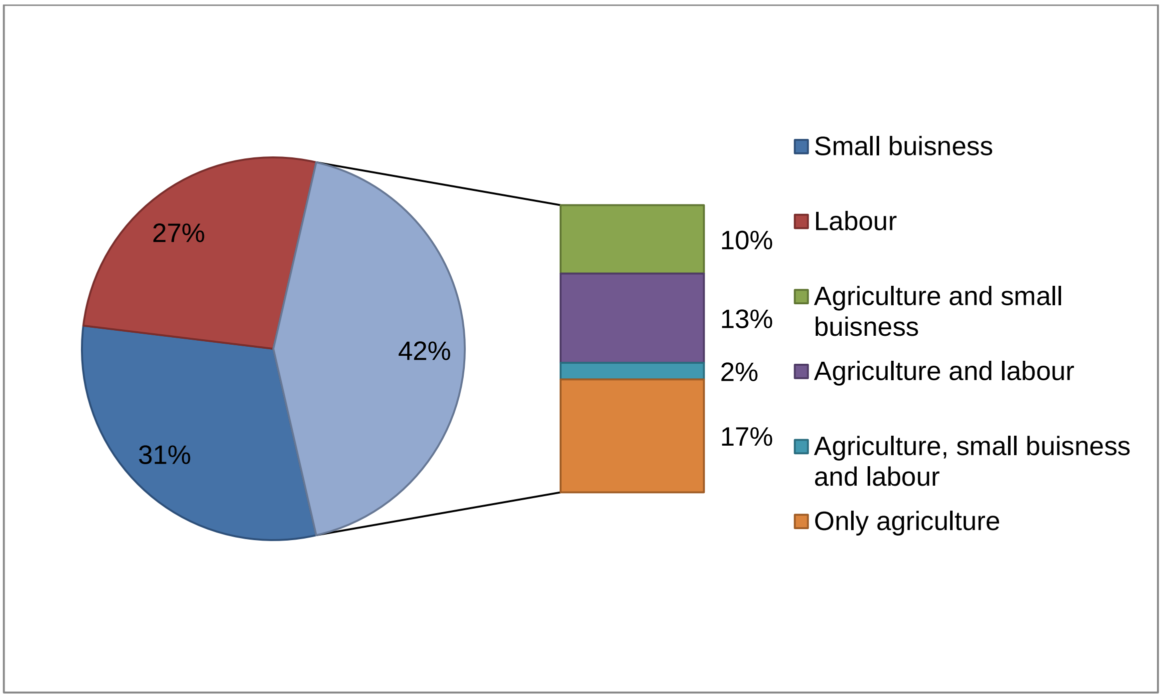

Figure 32: income sources

All further calculations in the master plan are based on the prospective population numbers and refer to demographic changes. The increase in population and the main issues announced in the master plan are to be dealt with by means of modernization and industrialization.

5 Discussion

5.1 Soil samples

The paddy soil analyses show that the soil is relatively low in total As (mean 9.3 mg/kg). The topsoil has lower concentrations than the root zones. The Pearson analyses of the topsoil and the root zone layer revealed quite different results: in the upper zone layer, no correlation was found between As and five heavy metals whereas the correlation in the root zone layer appears significant. This can be attributed to leaching processes at the soil/water interface.

The sequenced fractionation procedure was carried out to study the potentially bioavailable As in the soils (Figure 19) (Hoang Trang, Hahn 2015). To extract fraction F1 and F2, mild extractants were used ($(NH_4)_2SO_4$ and $(NH_4)HPO_4$) accompanied by shaking over 4 and 16 hours respectively). The extracted amount of F1 is very low: between below the detection limit and 0.6% of the total As content. The extracted As of the second fraction is between 3.6 and 12.9%. This would mean that the bioavailable amount of As is very low, which contradicts the results of the vegetable and rice analyses. As a matter of fact, the role of the different fractions in regard to the bioavailability of the As is not yet completely explained (Figure 33). Especially the fraction of As which is bonded to amorphous and poorly crystalline iron hydroxides (F3) seems to have different bonding characteristics. (Liu et al. 2015, Du et al. 2008, Tang et al. 2007) have found that the As content in F3 is not bioavailable, whereas Hsu came to contrary results. (Tufano et al. 2008, Liu et al. 2015) postulate a close relationship between the bioavailability of As of F3 and the oxidation state of the As. As(III) is much more mobile than As(V) and the reducing conditions in the paddy fields probably encourage the abundance of As(III). Thus in this study, F3 accounts for the bioavailable fractions, which means that half of the total As is bioavailable.

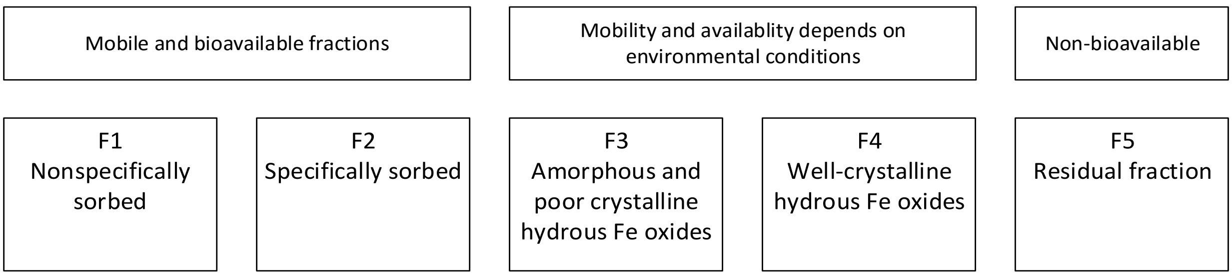

Figure 33: Bioavailability of the five fractions

These results should be considered bearing in mind the special conditions in the rice cultivation regions. The village of Dai Lam is situated in an agricultural area which has been used for rice cultivation for millenniums. In the Red River Delta, wet rice cultivation has been carried out for more than 5,000 years (Higham 1984), and it is a known fact that the soil characteristics are strongly influenced by the long-lasting cultivation. As a result, these soils are even considered an independent unit in soil classification

(IUSS Working Group WRB 2007). Farming activities such as plowing and puddling, flooding and draining, manuring, liming and fertilization have formed the soil’s characteristics which have been studied by international pedologists (Hao 2008, Zi-Tong 1983, Yang et al. 2005, Kirk 2004). The paddy soils are grouped into three categories, depending on their irrigation and moisture regime: oxidizing, oxidizing-reducing and reducing (Huang 2015). The paddy fields in the study area are flooded for about nine months a year and are considered as oxidizing-reducing, whereas the periods of reducing conditions predominate and are likely to have influenced the As mobilization regime over thousands of years.

5.2 Groundwater samples

A close look at the groundwater analyses and water use results indicates three main particularities, which are outlined below.

5.2.1 High arsenic concentrations

According to the investigations by Winckel et al (2011), the area of Dai Lam is located outside the zone of high risk of arsenic contamination in the groundwater. The highest concentrations in the southwest of the modern Red River course are linked to the presence of the lower Holocene aquifer and sediment layers high in organics (Figure 34). The geological setting of the area of Dai Lam is characterized by upper Pleistocene sediments, which indicate a minor arsenic risk. Nevertheless, the groundwater analyses in Dai Lam showed arsenic concentrations up to 106 µg/l with a mean value over all campaigns of 23 µg/l. In the study by Winckel et al., similar outlier values were found and explained by horizontal or vertical leaching effects from upper Holocene layers into the Pleistocene aquifers. The concentration of arsenic in groundwater is thus a matter of availability which depends on the presence of arsenic near the water-bearing sediment layers and the mobilization conditions. It has been shown that a series of factors influences arsenic mobilization (see chapter 2.4.2) such as the bonding minerals, pH and ORP conditions, the presence of other competing anions and organic matter.

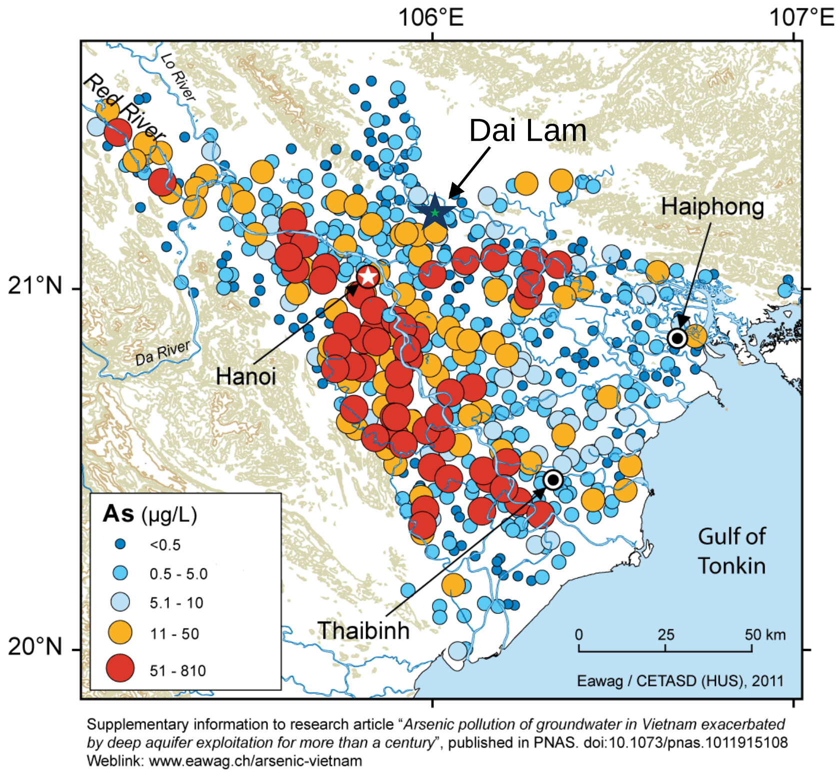

Figure 34: Arsenic concentration in the Red River groundwater (Winckel et al, 2010)

A comparison of Pleistocene and Holocene groundwater samples in Nam Du, near Hanoi has shown that both aquifers are affected by arsenic contamination (Norrman et al, 2006). Taking into account that the main source of the arsenic is located in the Holocene sediments, the contamination of both aquifers is a result of an interlaced sediment structure and complex mobilization. Therefore, the high concentrations in Dai Lam confirm previous studies.

5.2.2 Strong temporal and spatial variation

As described in chapter 4.2, the sampling campaigns showed that some wells have arsenic concentrations which are subject to high temporal and spatial variation (Figure 35). The period between the three main sampling campaigns (almost nine months) was too long to permit reliable assessment of the arsenic concentrations over time; monthly measurements were not feasible. However, this high variation raises a number of issues, which is why additional sampling was carried out in four wells in 2014.

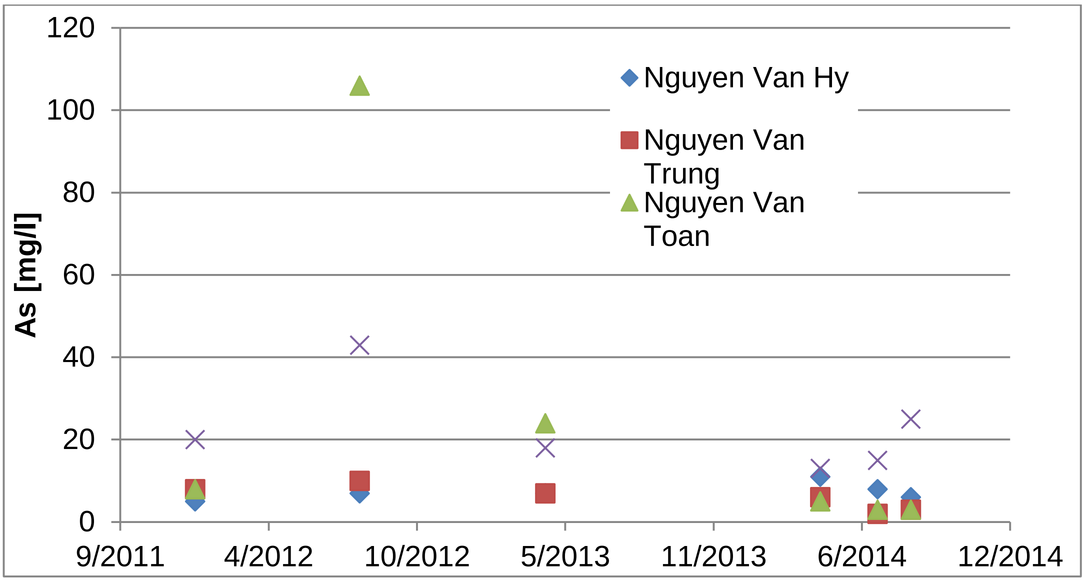

Figure 35: Temporal arsenic variation in four wells in Dai Lam

A variation analysis of eleven wells with three or more samplings was carried out. It was found that only three wells out of eleven had a variation coefficient smaller than 1 (Table 22), indicating high temporal variation, which is in line with other studies in Bangladesh and China (Cheng et al. 2005, Han et al. 2013).

Table 22: Coefficient of variation of arsenic in 14 wells, temporal variation. Wells 1–4 were sampled eight times (red), Wells 5–14 were sampled three times (blue), and Wells 15–20 (green) were sampled once or twice.

	Household name	Coefficient of variation	Number of samplings
1	Nguyễn Văn Hy	3.55	8
2	Nguyễn Văn Trung	1.98	8
3	**Nguyễn Văn Toan**	**0.6**	8
4	Tran Van Son	2.04	8
5	**Vũ Xuân Hiền**	**0.99**	3
6	Nguyễn Thị Tâm	1.03	3
7	**Trần Văn Giang**	**0.6**	3
8	Nguyễn Văn Vi	1.04	3
9	Nguyễn Văn Phúc	1.34	3
10	Nguyễn Văn Phương	1.18	3
11	Nguyễn Văn Nghi	1.01	3

The mobilization processes of As have been studied and discussed in several international publications and are described in chapter 2.4.2. The temporal and spatial variation of As concentrations is explained by specific local natural and anthropogenic conditions.

The phenomenon of temporal variation has already been reported by other authors in Spain (Mayorga 2013), Inner Mongolia (Guo 2013, Farooq 2011), India (Farooq 2011, Ramesh Kumar, Riyazuddin 2012), Bangladesh (Cheng et al. 2005) and China (Han et al. 2013, Yu 2014). The studies were carried out over several years in order to investigate whether the variations follow a seasonal trend and are linked to other parameter changes. The studies proved that wells with a high variability in their arsenic content are in shallow aquifers at a depth of less than 40 m, although only some wells (fewer than 20%) showed strikingly high variability. In some cases redox-sensitive constituents such as P, dissolved S and Mn varied in concert with the As values, but Fe couldn't be correlated (Cheng et al, 2005).

All authors have observed a cyclical, seasonal pattern of As variation in some wells, although there is evidence that some wells show different, non-rhythmical variations.

In the case of Dai Lam, no cyclic seasonal variation could be ascertained because consistent sampling was not carried out. However, the high variability observed is a result of many local natural and anthropogenic impacts leading to different mobilization, sorption and transport processes, which are described below.

The hydrological transport process is a key factor in the temporal and spatial variations. Vertical seepage of precipitation, irrigation and river water follows a complex pattern over the year and significantly affects the composition of the groundwater by means of dilution and sorption. Higher seepage rates have diluting effects and it was assumed that during the wet season the concentration of As and other components in the shallow aquifer are lower than in the dry season; this is seasonal pattern has in fact been demonstrated by some studies. Farooq et al. (2011) showed that some wells have decreasing As concentrations after the monsoon rainfalls in West Bengal and attribute this fact to dilution and oxidizing effects. Savarimuthu et al. (2006) described decreasing concentrations during and after monsoon in some wells. However, several studies show the opposite effect of increasing concentrations after the monsoon season (Ramesh Kumar, Riyazuddin 2012, Savarimuthu et al. 2006, Yokota 2001, Guo 2013). It is likely that As variation is linked to the variation of the groundwater level; one possible explanation is the flushing of soil and unsaturated zone after the dry season and/or a change of the ORP towards anoxic conditions, leading to the dissolution of HFO and thus to the scavenging of As and increasing concentrations of As (Cheng et al. 2005). Guo et al. (2013) postulated that the release of As in the groundwater is a result of restraining the dispersion of atmospheric O_2 because of irrigation activities.

In the RRD, the monsoon's main precipitation period begins in May and ends in August–September. Most of the precipitation water runs off and is drained into the receiving streams. The percolation and groundwater recharge of the upper aquifer is fast, the coefficient of permeability of sandy-loamy sediments being 0.02-1 m/d. It can be assumed that the percolating water only needs 1–4 weeks to reach the upper aquifer. The groundwater level reaches its maximum in September. The upper aquifer is also affected by seasonal temperature variations of 4–5°C with a minimum in February and a maximum from May to August (Giang 2014). In this regard, it seems plausible that in some wells the As concentrations rise in the wet summer season and drop in the dry winter.

However, apart from the climatic impact, the variation of the As concentration in the groundwater in Dai Lam is likely also a result of anthropogenic activities (Figure 37) and the resulting input of OM, nitrogen, phosphate and sulfate species. Land use in the study area is dominated by paddies. The special conditions of this kind of agriculture imply seasonal variations of the physicochemical properties of the topsoil and soil water. There are two cultivation periods every year (see Figure 36): one in spring and the other in summer/autumn. The seeds are sown between December 10 and 16. The rice seeds germinate after 10–15 days in concrete beds or after 25 days in the paddies. Two weeks after germination, the small seedlings are bedded out. Two and a half months after seeding, the plants begin to bloom, and after 110–120 days (spring) or 95–110 days (summer/autumn), harvest begins.

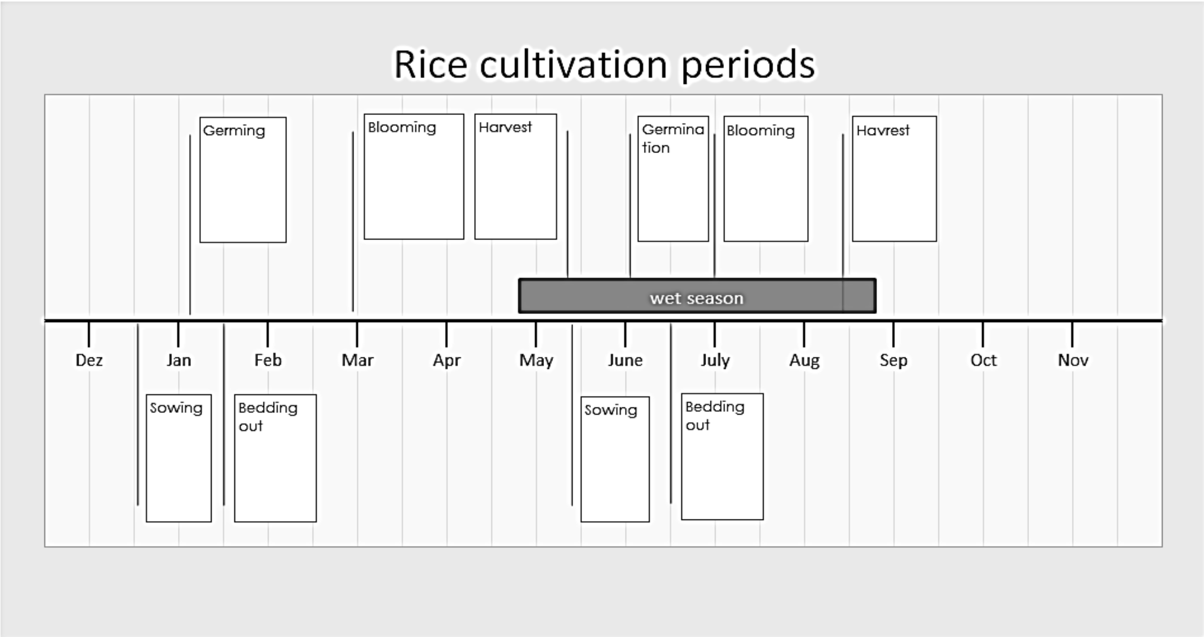

Figure 36: Rice cultivation periods in RRD

The climatic and agricultural activities and their possible effect on the As specifiation are shown in Figure 37. During the first few weeks of plant growth, water demand is high. In January, when the seedlings are small, in the area of Dai Lam the irrigation pumps are in operation some 70 hours within 10 days, amounting to 228 l/m^2 (corresponding to 13% of the annual precipitation in the RRD). The irrigation water is pumped from the river and in the wet season the water in the paddies is rainwater. Both river water (ICEM, 2007) and rainwater have negligible As levels and it must be assumed that any As which occurs in the aquifer or in plants originates from the soil. Four weeks before harvest, the paddies are drained and the fields remain dry from September until December. In this time, the conditions in the soils are aerobic. Under aerobic conditions, As(V) is the predominant species and it is sorbed to soil minerals like iron, manganese and aluminum (hydr)oxides (Goldberg 2002b). After flooding, the conditions turn anaerobic and a microbially induced reductive dissolution of the Fe and Mn (hydr)oxides (Cummings 1999) leads to the release of As into the soil water, where it is reduced to As(III). During the growth of the rice plants and the development of the root aerenchyma, micro-aeration leads to aerobic conditions in the rhizosphere (Li

2013b) and the precipitation of Fe and Mn (hydr)oxides on the roots (Garnier et al. 2010). These Fe/Mn plaques are typical in the paddies of the RRD and As(V)is sorbed on the plaques. The formation of Fe/Mn plaques increases over the cultivation period (Mei et al. 2012) and it was shown that most of the sorbed As is composed mainly of As(V) (Voegelin 2007). The formation of Fe/Mn plaques was observed to decrease at the end of the growing period (Schmidt 2011). Hence, the wet cultivation period is characterized by anaerobic conditions in the soil water and As is predominantly present in its reduced mobile species As(III). The water subsequently percolates through the sediment layers. Reducing conditions within the percolation lead to the further release of As from the sediments and the enrichment of As in the groundwater.

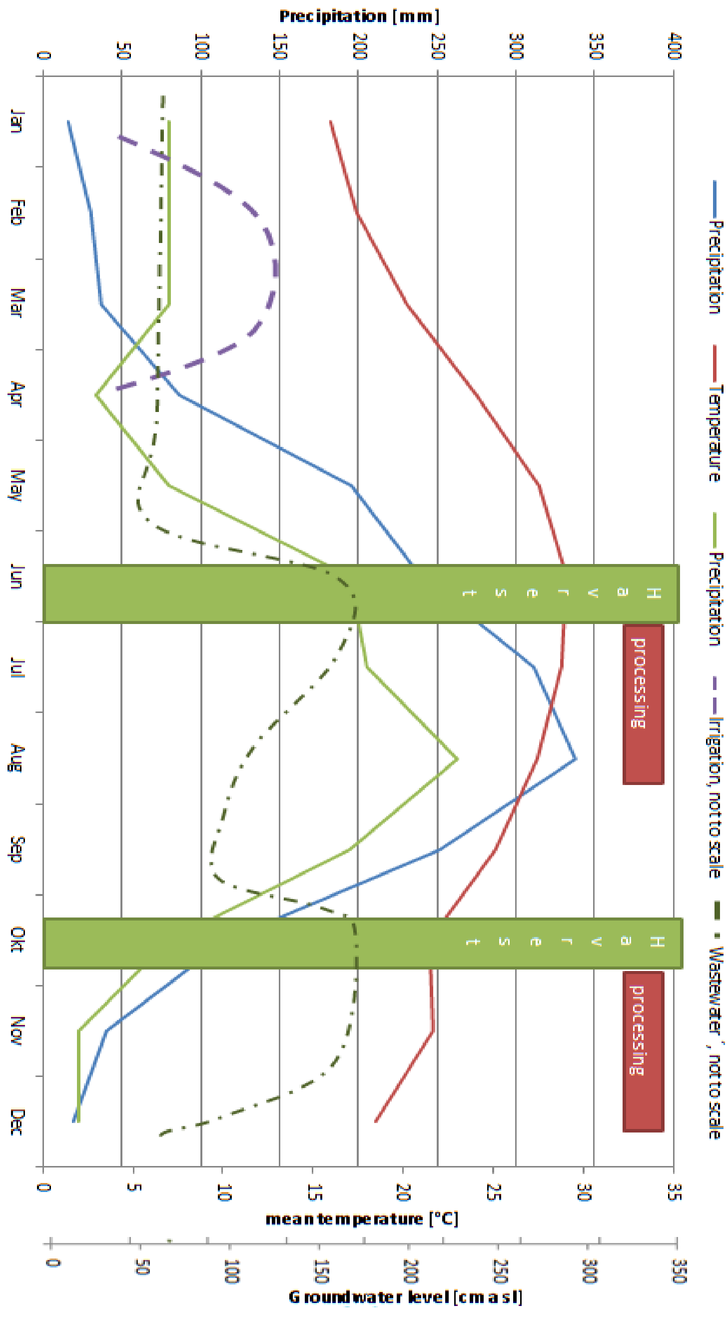

Figure 37: Processes and conditions affecting arsenic specification and mobilization

After the rice harvest, residual rice straw and roots remain in the fields and undergo anaerobic microbial decomposition in the flooded paddies (Farooq et al. 2010).

Reducing conditions lead to the mobilization of As and to vertical transport to the aquifer. Furthermore, these processes produce organic acids which act as electron donors for microbiological processes in the unsaturated zone, which may provoke the dissolution of iron hydroxides and the release of adsorbed As (Farooq 2011). Additionally to the above-mentioned factors, rice cultivation requires the application of fertilizers, and the mineral components can have very different effects on As speciation. (Acharyya et al. 1999, Anawar 2011, Chowdhury et al. 2000) reported that agricultural fertilizers and organic wastes may enhance the mobilization of As by ion exchange with phosphorus derived from fertilizers. Nitrate originating from fertilizers leads to the oxidation of Fe(II) and to the generation of HFO, which decreases the arsenic content in the groundwater (Mayorga 2013). On the other hand, nitrate acts as electron acceptor (with a redox potential between O_2 and Fe(III)) and may thus imply reducing conditions.

Dissolved carbonates can displace As from sorption places and thus lead to the mobilization of As (Appelo et al. 2002), while bicarbonate can even extract As from the sediment under oxic and anoxic conditions (Anawar 2003). In Dai Lam, fertilizer application is limited to the cultivation period and is thus a temporary varying factor.

The effects of paddy agriculture and natural water level variations are increased or at least overlaid by the addition of wastewater to the irrigation/drainage systems. As mentioned above, Dai Lam is a traditional food processing craft village. Until 2012, the wastewater of the whole village was drained untreated into a pond and subsequently entered the irrigation system (0). One specific feature of food processing craft villages is the high content of COD, NO_3^- and NH_4^+ and their high variability in wastewater quantity and quality over the year (4.2.3). Although the wastewater analyses by the University of Hanover (Meier, 2014) showed very different results over the three measuring campaigns, assuming a mean wastewater quantity and quality, the annual effluent of the village calculated from the three campaigns is 137 t of COD, 0.14 t of NO_3^-, and 23 t of NH_4^+, which are drained into the pond. These processes contribute substantially to the anaerobic and reducing conditions in the uppermost sediment layers which are the main sources of As in the groundwater.

The production of wastewater in the village of Dai Lam varies in quantity and quality. After both annual harvests, the processing activities in the village increase, causing the amount and composition of wastewater to rise sharply in parallel. The wastewater of the whole village is collected in the canal system, but no treatment facility was installed until 2012 and the wastewater was collected in a pond before being drained into the irrigation system. Only very few studies have been published on the effects of sewage sludge application, but (Karczewska 2013) showed that spreading sewage sludge on contaminated soils (to improve the natural attenuation) increased the bioavailability of As. The reasons are sorption/desorption mechanisms and chelating reactions caused by the application of low-molecular weight compounds.

Equally, very few studies have studied the effect of untreated organic wastewater on the presence of arsenic in groundwater, although recent studies have investigated the role of agricultural fertilizers and OM in the occurrence of As in the shallow groundwater in Bangladesh (Anawar 2011, Anawar 2013).

Similar to OM and its effect on As mobilization, microbial sulfate reductions have rarely been discussed as a relevant factor for As release. Recent studies have shown that microbial sulfate reduction has been underestimated in this context (Burton 2007, Burton 2013). Dithioarsenate and monothioarsenate have been proved to be the major As species under sulfate-reducing conditions. Both substances are poorly adsorbing and may play an important role in the formation of mobilized As species under sulfate-reducing conditions (Burton 2013).

The spatial variation of As in the wells of Dai Lam is a phenomenon which is also observed in other sites in Vietnam, and is a result of the small-scale geological, hydrogeological and hydrological setting. OM in peat lenses may provoke sharp differences between sampling sites that are only a few meters apart from each other. Silty-clayish sediments have higher adsoption abilities than sandy sediments, while one of the characteristics of deltaic lacrustine sediments is that the sedimentary composition is variable (van Geen 2003, McArthur 2001b, Eiche 2008). (Larsen 2008) explained the spatial variability of As concentrations by hydraulic properties of the overlying layers. Thicker clay layers can inhibit the vertical transport of electron acceptors such as O_2, NO_3^- and SO_4^{2-}, hence in aquifers beneath thick clay layers conditions are reducing, Fe(hydr)oxides are reduced, and As is released into the aquifer. In addition, Larsen et al. (2008) found that no O_2, NO_3^- or SO_4^{2-} was present in the aquifer beneath a channel, assuming that this occurred because of reducing processes in the bottom of the channel. The interface layer between surface water bodies and the pedosphere is characterized by complex biochemical processes, which may also have strong effects on the redox conditions (Sophocleous 2002, Larsen 2008). This indicates the clear impact of surface water canals and ponds on subterranean water conditions.

5.2.3 Weak correlation between measured parameters

In order to analyze the extent to which the individual parameters correlate with the arsenic concentration, a Pearson product-moment correlation was carried out. The results are presented in Table 23. The correlation analyses show that the sampling campaign in 2011 reveals a correlation between NH_4^+, ORP and TOC (red letters), while the following campaigns in 2012 and 2013 showed no significant correlation between arsenic and the appropriate parameters. Hence, the temporal and spatial variation of the arsenic concentration cannot be explained by the change to the redox conditions, the degree of pollution, or the electrical conductivity (see also the further discussion later).

Han et al. (2013) found a link between As variability and HCO_3^{-}. In periods of strong changes in the As concentrations, the HCO_3^- concentrations shows similar amplitudes.

Table 23: p-test of arsenic and appropriate parameters

		NH_4^+	ORP	EC	DO	TOC	Mn
2011	Correlation coefficient	–0.495	–0.455	–0.172	0.108	–0.517	–0.57
	p-value	0.0264	0.0437	0.468	0.651	0.019	0.004
2012	Correlation coefficient	–0.158	0.0741	–0.0991	–0.276	–0.083	–0.416
	p-value	0.451	0.725	0.637	0.181	0.692	0.068
2013	Correlation coefficient	–0.422	–0.102	–0.115	–0.515	0.392	–0.491
	p-value	0.0808	0.678	0.638	0.023	0.097	0.033

Similar to the situation described above, no significant correlation was observed between arsenic and manganese.

The sampling campaign in summer 2012 showed by far the highest concentrations, but also the widest variation in the concentration: the mean was 44.1 µg/l, and the standard deviation was 33.2 µg/l, whereas the mean concentrations in winter 2011 and spring 2013 were 10 and 17 µg/l, respectively.

Although the variation of the arsenic concentration at each sampling campaign was high, other parameters like dissolved oxygen, NH_4^+ and TOC were relatively constant, but showed the typical values of an anoxic, agriculturally affected shallow aquifer Table 24).

Table 24: Mean results of water analyses

Year	T	pH	DO	EC	ORP	SO_4^{2-}	NH_4^+	TOC	As	Fe	Mn	Coliform
	°C		[mg/l]	[µS/cm]	[mV]	[mg/l]	[mg/l]	[mg/l]	[µg/l]	[mg/l]	[mg/l]	
2011	25.5	6.8	1.2	785	–139	41.3	5.43	14.87	10.5	4.8	0.15	4.0
SD	0.3	0.1	0.8	324	31	36.2	1.76	4.99	8.6	3.2	0.12	
2012	25.9	7.1	1.3	765	–149	31.8	6.55	7.15	44.2	32.8	1.01	5.5
SD	0.5	0.2	1.5	332	32	23.0	3.38	4.63	32.3	20.9	0.46	
2013	23.8	6.9	1.8	633	–127	28.1	5.01	7.43	17.2	16.5	0.45	10.1
SD	1.0	0.3	0.8	180	36	41.9	3.59	3.63	16.5	11.7	0.42	

5.3 Wastewater and sewage sludge

The sewer system in the village of Dai Lam was built in 2006 and consists of small covered canals in the streets which collect the wastewater from the households and drain it into an open canal conducting the water to the irrigation system (see Figure 21). The wastewater in the village of Dai Lam is characterized in different ways by the rural and processing activities. The diurnal variation of the wastewater quantities shows two peaks of wastewater production: one in the early morning from 4 to 7 am, and the other in the afternoon from 3 to 6 pm. The first peak is a result of the processing activities in the crafting households while the later peak is caused by everyday activities in the families. Over the year, the high, seasonal variation of the wastewater amount has been observed (Meyer, 2015), which can be explained by the two rice harvests and the subsequent processing activities in the villages which consume a lot of water. In addition to the quantity of wastewater, the effluent has a high amount of organic matter (COD), which is typical of a food processing craft village. The As level in the wastewater is low to moderate and is approximately the same as the As level of the groundwater. The removal efficiency of As in the SBR pilot plant is 71%. Although the SBR is not known to be an appropriate process for As removal, the oxidizing conditions provide good pretreatment because of increased precipitation (Andrianisa et al. 2006). However, the As content is accumulated in the sewage sludge. In Vietnam, sewage sludge management is regulated by TCVN 5298:1995 ‘General requirements for the use of wastewaters and their sludge for watering and fertilizing purposes’. However, it is weakly enforced and especially in the rural areas most sludge remains untreated and is reused as fertilizer in the paddies.

The analyses of the sewage sludge showed that the As contamination of the samples is moderate at 30 mg/kg. Thus, different point of views surround risk assessment of the reuse of the sludge as fertilizer: possible As contamination may provoke As sinks whereas the high amount of organic matter (which affects the physicochemical properties in the soil) enhances the mobilization of As.

5.4 Pig manure

The As concentration in the manure samples in Dai Lam showed a strong correlation between the food source and the As content in the manure. Evidently the soaked rice, which is traditionally used as a food source in the villages, contains much higher As values than the manure of animals fed with industrial food. The manure plays an important role as fertilizer in the paddies, and in Dai Lam part of the manure is sold to junk dealers to be used as fertilizer elsewhere. Though the number of samples in this study doesn’t constitute a reliable data pool, the relevance of the results must be highlighted and should be studied in detail in future, because the reuse of manure containing might As produce unexpected As and heavy metal sinks in rural regions.

Chinese studies have proved that swine meat production in several regions in China led to As levels of up to 2.7–57.2 t/ha/a in the Beijing area (Li, Chen 2005). Due to the weak data pool of this study, further investigations are required into the possible effects of meat production in the Red River Delta on the As exposure risk. So far, no reliable analyses of the heavy metals in pig manure have been published. In addition, the effect of As-containing manure on paddy fields has not been quantified.

5.5 Daily exposure to As from dietary intake

In chapter 4.5.2, the As concentrations of edible vegetables and other plants are presented. Although the amount of data is too small to be representative, the data proved the significant As uptake in plants. The concentration of As varies and depends on the kind of plant, its lifetime, the growing period, As concentration in the soil, and mobilization factors, which are strongly linked to climatic conditions and anthropogenic activities (chapter 5.2).

In this section, the villagers' possible daily intake of As is estimated (Table 25). Because the main agricultural product of Dai Lam is rice and pork, only limited data on vegetables, fruit and other goods could be gathered in the village itself. Thus, the estimate relies on the data gathered in this study and data from the literature review. In general, the range of As concentrations in food samples is very wide. The highest concentrations are found in seaweed and seafood (Figure 38).

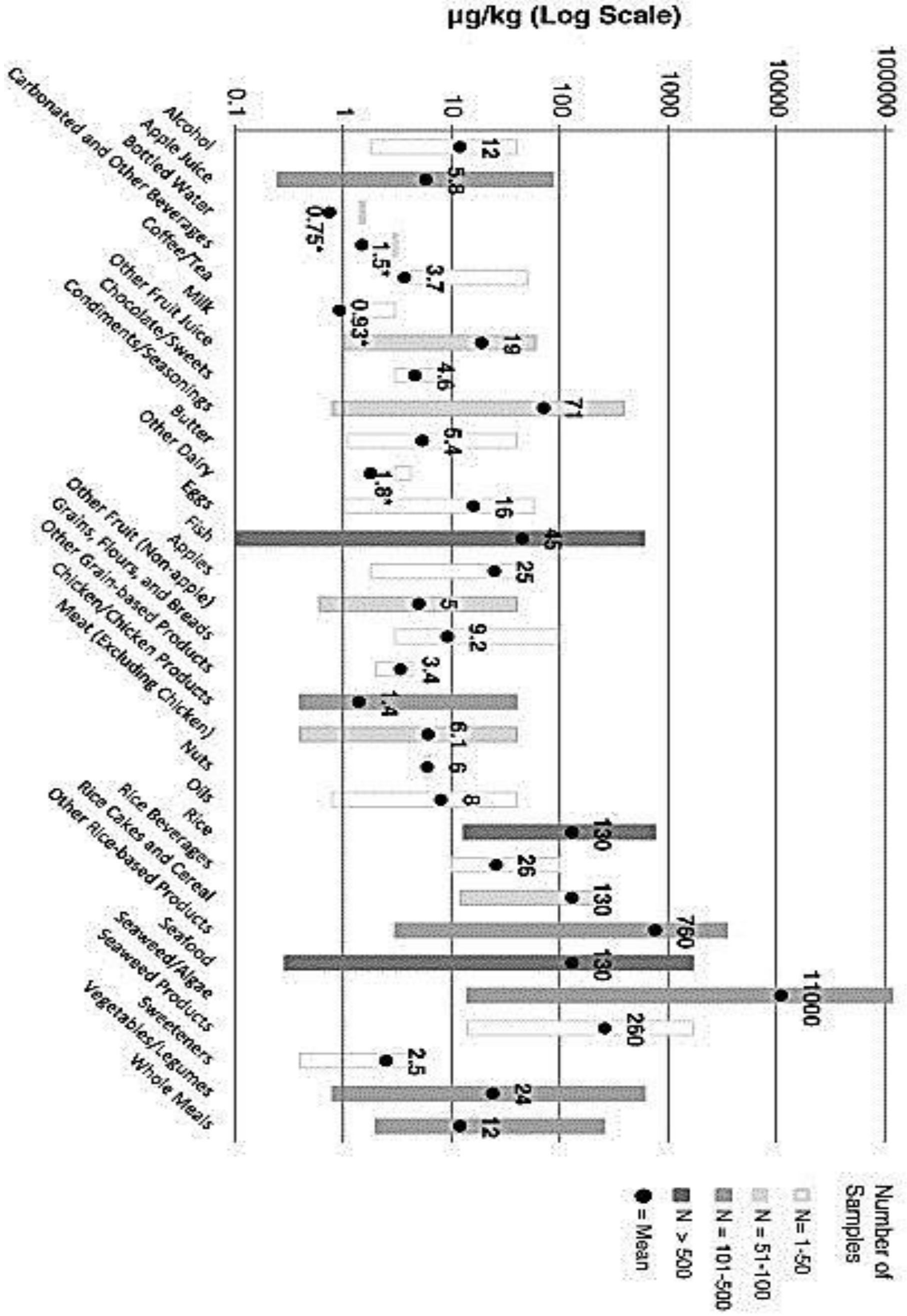

Figure 38: Mean total inorganic arsenic in foods and beverages (µg/kg unless otherwise noted). Note: * Because all or most of the samples were below the limit of detection (LOD), half the LOD was used. Thus, means are below the measured range (source: Lynch et al. 2014)

Rice:
The As content in the rice samples reveals high variability. In four grain samples out of eleven, the concentration is lower than the detection limit while five samples exceed the current Chinese threshold for As in rice of 0.15 mg/kg. The results of rice grains obtained in this study correspond to other results presented in chapter 2.5.5.2. The mean value of rice grains is 0.15 mg/kg and the maximum 0.59 mg/kg, showing that the intake of local rice might pose a health risk to the villagers. However, the human risk posed by the rice grains depends on As speciation as well as total grain concentration. In this study, As species were not determined, but other studies (Norton et al. 2010) showed that the level of As_i in rice varies between 60 and 95% (Halder et al. 2014). It is thus assumed that the As content in the rice at the study site largely consists of inorganic As. Like the rice grains, the rice plants also reveal high variation within their As concentration. Twenty-six samples of rice plants were analyzed. Depending on the material, leaves and stems were assessed separately or as a mixed sample. The mean of all 26 samples was 4.09 mg/kg dw and the standard deviation was 2.4 mg/kg, which is in line with other studies. It should be mentioned that the samples of the leaves have relevant higher As concentrations than the stems, which may indicate either the enhanced accumulation of As in the leaves, dusty accumulation on leaves which is resistant to normal washing and rinsing, or direct uptake through the leaf's surface, which would be out of step with other studies, which postulate uptake via the roots (Zhao et al. 2009, Ali et al. 2009, Abedin et al. 2002b) .
Vegetables (leaf vegetables and non-leaf vegetables):
Vegetables are subdivided into leaf vegetables and non-leaf vegetables. In traditional Vietnamese cuisine, the amount of leaf vegetables exceeds the amount of non-leaf vegetables. Leaf vegetables like spinach and cabbage accumulate higher As concentrations than other vegetables such as kohlrabi and tomato. In Dai Lam, the total possible As concentration in fresh plants may be as high as 0.49 mg/kg in white cabbage or 0.16 mg/kg in water spinach (chapter 4.5.2), which are both very common components of the daily diet. These findings are in keeping with other studies (Baig, Kazi 2012, Das et al. 2004, Wang et al. 2013). In comparison to European leaf vegetables, the concentrations stated are ten times higher than documented European samples. In the study area, only limited non-leafy vegetables were sampled (kohlrabi with As content of <0.002–0.025 mg/kg). In northern Thailand, tomato and chili peppers contained 0.035–0.074 mg/kg and 0.062–0.126 mg/kg, respectively (Thaharn 2014).
Meat:
The meat analyses in Dai Lam (chapter 4.5.4) revealed that the As concentration depends highly on the feeding habits. The pigs that were fed with local rice accumulated higher As concentrations in the liver (0.42 and 0.79 mg/kg) than the pig which wa fed with industrial food (0.01 mg/kg). The meat sample had a concentration of 0.07 mg/kg. In the poultry samples, the concentration in the meat was below the detection limit while the liver contained 0.176 mg/kg on average. Because the data are not sufficient to be considered reliable, other data are consulted for this review. In the EFSA report, European pork and meat samples had an average As concentration of 0.007 mg/kg for grassland-raised animals and 0.014 mg/kg in the edible offal of game animals. A Taiwanese study found an average As concentration in meat of 0.005-0.03 mg/kg, pig and poultry samples having higher concentrations than other meat samples (Chen et al. 2013). Lynch (2014) referred to As concentrations in poultry samples of <0.0017–0.16 mg/kg and in other meat samples 0.005–0.075 mg/kg. These results revealed that especially the offal of the animal livestock in Dai Lam may have high As concentration if the feed

consists mainly of local rice. The content of inorganic As varies between 30 and 66% (Lynch et al. 2014, Phan et al. 2013)
Fruit:
No fruit was cultivated in the study area during the investigations; moreover, the data pool for As in fruit is weak. In Europe, the As concentration in citrus, pome and stone fruit is 0.043–0.005 mg/kg (EFSA 2014). In Thailand, banana samples had 0.002 mg/kg (Nookabkaew et al. 2013a) and in Hainan Island, China, 0.0006–0.50 mg/kg was detected in mangos (Liao 2014). In other studies addressing human exposure to As, fruit is not usually considered because of insufficient data (Signes-Pastor et al. 2008).
Fish:
In Dai Lam, four fish farms are run with different fish. For this study, the liver and meat of three northern snakehead fish (*channa argus*) were analyzed, but the results were below the detection limit. However, the potential risk from fish consumption in Dai Lam should not be ignored, because snakeheads are carnivore fish which are fed with industrial food. The results from a comparable study area with similar As concentration in the soil in the Pearl River Delta in China (Cheng et al. 2013) showed that As concentrations in fish depend on the fish species. Their nutrition behavior, living area (water or mud) and age have a significant impact on As accumulation in the meat. Largemouth bass samples have by far the highest As level (2.23 ± 0.57 mg/kg ww), which was one order of magnitude higher than the other fish samples. The other fish samples from this study (Cheng et al. 2013) had concentrations between 0.12 ± 0.08 and 0.47 ± 0.08 mg/kg ww, which is endorsed by the results achieved by (Cheung et al. 2008). The comprehensive study by (Lynch et al. 2014) found a mean concentration of 0.045 mg/kg in fish. Nevertheless it must be taken into account that in fish samples up to 90% of the As content is made up of the non-toxic arsenobetaine (Kar et al. 2011, Lin, Liao 2008, Liang et al. 2011) and that the inorganic As species in fish only accounts for between 3.5% and 10% of the total As (Lynch et al. 2014, Phan et al. 2013).
Eggs and milk:
In Europe, the average As content in milk and eggs is 0.0016 mg/l and 0.0584 mg/kg, respectively. Though the consumption of these products is much lower in Vietnam than in Europe, the contribution to the As intake through these products has to be considered because the concentration of As might be much higher. To date, no reliable data are available about the As specification in eggs.
Tofu
There are only very limited data available on the As concentration in tofu, even though it is a common element of Asian cuisine. The EFSA report (2014) specified an As concentration of 0.013–0.02mg/kg. No information is available on the specification of As in tofu.
Wheat:
No wheat is cultivated in the study area, but bread made from wheat became an important part of the Vietnamese diet during French colonization. As uptake in wheat is, compared to rice plants, weak. The EFSA report states an average As concentration in wheat of 0.017 mg/kg in Europe, and the same value was ascertained by Lynch (2014), the majority of which is inorganic.

Other grains:
Grains such as corn, barley, rye, spelt, buckwheat, millet and oat have As levels of 0.011–0.028 (EFSA, 2014)
Seafood:
Seafood is recognized as one of the major sources of As in the human diet. However, the concentration depends highly on the kind of species. In the EFSA report, some cephalopods showed the lowest mean As concentration, whereas mollusks had the highest concentration (cockles). Thus, the concentrations found in seafood in Europe vary between 0.001 and 0.13 mg/kg. The comprehensive study by (Lynch et al. 2014) revealed a mean concentration of 0.13 mg/kg in seafood (0.02–0.8 mg/kg). The golden apple snails which were analyzed in this study contained up to 1.01 mg/kg. In contrast to other seafood, the As in gastropods consists of inorganic trivalent As (chapter 2.5.7) rather than the relatively harmless arsenobetaine.
Sauces:
In Vietnamese cuisine, fish sauce is a common condiment used like soy sauce or Maggi in Germany. The raw material of fish sauce is salt and fish (ratio 3:1), which is fermented for at least one year before being filtered and bottled. Mostly oyster, shellfish, shrimps and other small fish are used as well as residues from other fish-processing activities. Different fish sauce samples from Thailand and Vietnam were investigated by (Rodriguez 2009). The As content in the samples varied from 0.69 to 2.75 mg/l, whereas the average arsenobetaine level was 89%.
Oils:
No oil is cultivated or processed in the study area. However, oil is widely used in Vietnamese kitchens for frying and cooking. In Europe, the average As concentration is 0.008–0.023 mg/kg. Lynch found 0.013 mg/kg, of which 0.008 mg/kg is inorganic species.

Table 25: Average diet in Vietnam 2009 (National Institute of Nutrition (2009–2010)) and potential As intake

Average diet	Potential As intake	Average diet	Potential As intake
375 g/d rice	0–0.074 mg/d	20 g/d tofu	0.00025–0.00038 mg/d
160 g/d leafy vegetables	0–0.078 mg/d	20 g/d wheat	0.0002–0.0005 mg/d
84 g meat products	0.006–0.066 mg/d	17 g/d other cereals	0–0.0006 mg/d
70 g/d fruit	0–0.0035 mg/kg	15 g/d sauces	0.01–0.041 mg/d
70 g/d fish	0–0.033 (0.156) mg/d	10 g/ seafood	0.001–0.01 mg/d
30 g/d vegetables	0–0.0126 mg/kg	8 g/d oil and fat	0–0.0018
25 g/d eggs/milk	0–0.034 mg/kg	10 g/d others	

Figure 39 depicts the ranges and possible contributions of the different aliments to daily As intake (without water). The biggest contributor to As intake is rice, followed by leafy vegetables and meat products. In grains and other plant products, the level of inorganic As is much higher than the less toxic organic compounds. Based on these assumptions, the minimum daily As intake is 0.027 and the maximum is 0.42 mg/d, which would exceed the PTWI of a person weighing 75 kg (0.15 mg/d) as well as the Codex Alimentarius threshold of 0.2 mg/d (As_{in}) and 0.3 mg/d (As_{tot}) (chapter 2.3.2).

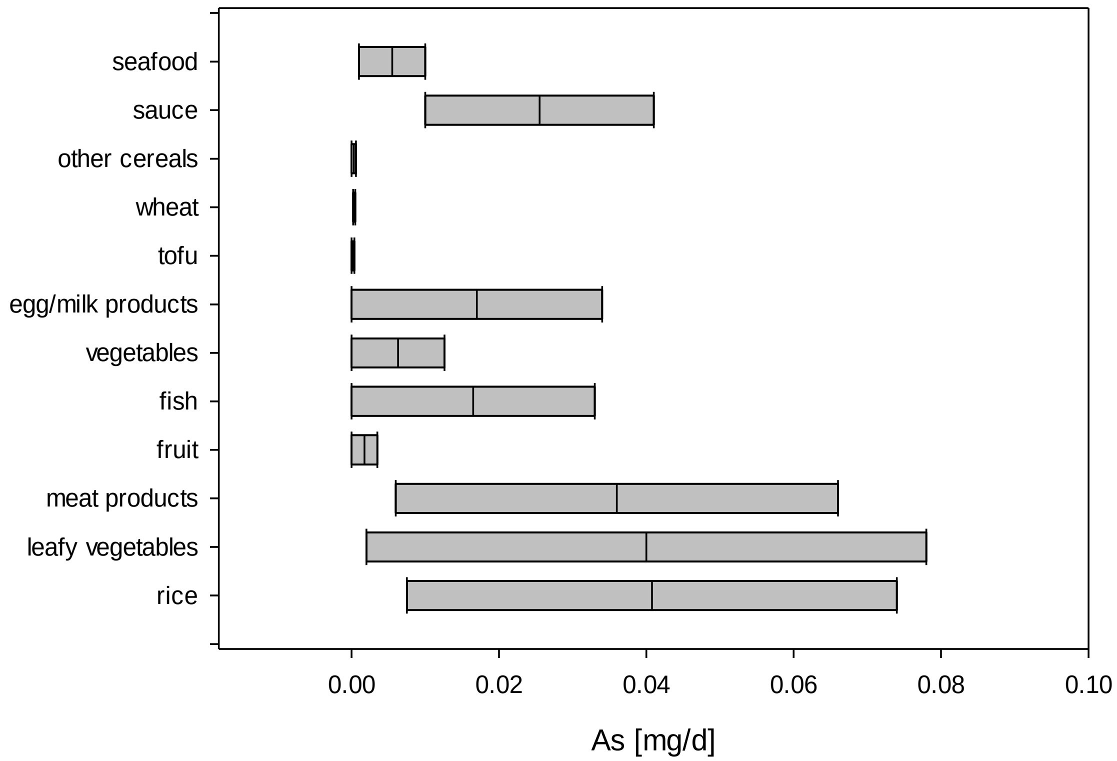

Figure 39: Ranges of potential daily As intake

As intake depends closely on the kind of food. Offal, leaf vegetables and rice potentially contain the highest As concentrations. Villagers and poorer people bear the highest risks of high As intake because their diet contains more rice, vegetables, offal and GAS, which have high As concentrations; moreover, fewer poor people are connected to the public water supply and rely on the shallow aquifer for their drinking water.

5.6 Effects of land and water use on water quality and public health

More than 80% of the households in Dai Lam are connected to the public water supply installed in 2008. The capacity of the waterworks is 180 m³/d, which is not sufficient for the village's demand. Low water pressure and financial reasons have led to the ongoing operation of the tube wells in domestic households. The interview campaign and our own measurements in 2012 indicated that two thirds of the water supply in Dai Lam originates from tube wells. Most households are aware of the risks of the tube well water and do not use it for cooking and drinking. According to the villagers, the tube well water is mostly used for cleaning, animal husbandry and production, with only a few people using it for cooking and drinking, most of the consumers using sand filters before uptake. However, the uncontrolled withdrawal of wastewater from the upper aquifer is a supplementary intervention in the hydrogeological setting and may lead to the further transport of As into the lower Pleistocene aquifer.

The quantity and quality of wastewater production in the village is typical of craft villages and is characterized by a varying load of contaminants and diurnal and seasonal fluctuation which is mostly a result of rice processing. Like most Vietnamese craft villages, the wastewater is drained untreated into the irrigation system, which leads to enhanced mobilization processes in the surrounding paddies and thus to the possible transport of As to the shallow aquifer. In the case of high As or heavy metal concentrations in the wastewater, the development of sinks are likely.

Although the area of Dai Lam is located on the northern rim of the Red River Delta, whose groundwater is reported (Winkel et al. 2011) to be only little contaminated by As in the aquifer, this study showed higher arsenic concentrations in the groundwater than expected. Furthermore, the variation of arsenic in the shallow aquifer is affected by strong amplitudes during the monsoon in August and September, which hampers the countermeasures and increases the human risk.

Concluding the well water analysis results, it can be inferred that the Holocene aquifer in Dai Lam, which is located at 18–22 m a.s.l., is clearly affected by agricultural activities, which is also confirmed by the occurrence of coliforms.

The landscape has been profoundly shaped by rural end economic activities over thousands of years. The Đông Sơn culture (1000 BC to 100 AC) introduced the widespread wet rice cultivation and the development of the irrigation system, which was also used as a transportation system (Taylor 1991), and which is still characteristic for the current landscape in the region.

French colonialization changed the transportation system from the water to overland. The socialist era introduced collectivization and increased industrialization, which resulted in an undersupply of staple food and to environmental pollution near the industrial areas.

Since the introduction of the Doi Moi policy, agricultural productivity has improved. Besides the higher efficiency of labor (due to increased responsibilities), improved irrigation systems and the application of fertilizer are the main reasons for the increased harvests. However, water use in the agricultural sector rose by 70% from 1980 to 2000. At the same time, water availability dropped due to huge hydroelectricity projects in China. Despite all the upheavals in land use patterns, production in the craft villages and traditional rice cultivation in paddies have continued.

5.7 Against the background of the transition economy

On January 25, 2014, the Prime Minister signed Decision 198/QD-TTg approving the overall master plan for the socio-economic development of the key northern economic center by 2020 and providing a roadmap until 2030.

Bac Ninh belongs to the ‘northern key economic zone’ which includes seven provinces (Hanoi, Hai Phong, Quang Ninh, Hai Duong, Hung Yen, Vinh Phuc and Bac Ninh). Because of its current economic status, this zone is supposed to be a leading zone in the rapid, sustainable socio-economic development of Vietnam (‘Master plan for the socio-economic development of Northern key economic center to 2020 and an orientation toward 2030 – Decision 198/QD-TTg').

In the last decade, Bac Ninh has undergone outstanding economic and social change, and this trend is set to continue (Table 26). In 2003, only 2,931 workers were employed in industrial enterprises, which predominantly were allocated to domestic enterprises. By 2012, 117,445 workers were occupied in industrial enterprises, and by the end of 2015, this number will rise to 180,000 due to several big investment projects. GDP per capita, which was US$1,488 in 2003, rose to US$3,500 in 2015, and is predicted to reach US$5,500 in 2020 and US$11,000 in 2030. This development is attracting many workers from other regions of Vietnam, where economic progress is weaker, and therefore the population growth of Bac Ninh is far above average (Dung 2014).

Table 26: Economic data of the northern key economic center (Socialist Republic of Vietnam, the Prime Minister 2014)

	2011–2015	2016–2020	2021–2030
GDP growth rate	7.5%	9%	8.7%
GDP per capita	US$ 3,200–3,500	US$ 5,500	US$ 10,500–12,000
GDP ratio of agriculture, fisheries and forestry	7.7%	5.5%	2.2%
GDP ratio of industry and construction	48.3%	49.1%	47.8%
GDP ratio of service	44%	45.4%	50%
Population growth	1.2–1.3%	1.1–1.2%	1–1.1%
Rate of urbanization	40–45%	50–57%	60–70%

The population of the seven provinces of the northern key economic zone will increase by 3.6 million people within twenty years and the number of persons living in urban areas will double. New residential areas need to be developed; for example, in Bac Ninh seven new building projects near industrial zones with a total area of 196,510 m^2 are being developed and built. Furthermore, several urban service centers with cultural monuments, social welfare units and other types of social infrastructure are being developed, which are allocated to the major industrial zones. New roads, transport media and infrastructure will be planned and built by 2020. The economic research institute *Prognos* states that the Vietnamese middle class is growing faster than in any other country, and so this trend will shortly be visible in the northern key economic zone. Although these projections imply a positive development for the region, the adverse impact on the environment will be significant.

The growth of the industrial and urban centers will lead to the intensified exploitation of the Pleistocene groundwater and to the increased production of wastewater. The greater use of the groundwater can lead to an accumulation of As from the upper aquifer due to changing hydrological patterns. The problem of increased wastewater production is far from being solved and may enhance the dissolution processes in the soil.

Urbanization, industrialization and increasing forestry are leading to the depletion of arable land. The government intends to maintain the high rice production, even though scientists object that intensive rice cultivation has adverse environmental effects (e.g. Professor Vo Tong Xuan, Acting Rector of Nam Can Tho University). The Mekong Delta, after hundreds of years of wet rice cultivation, “which produces about half of Vietnam’s rice, is showing signs of environmental stress. The earth dykes that were built to keep seasonal floods from inundating the rice paddies prevent the Mekong river’s alluvial floodwaters from bringing nutrients to the delta’s soil”[8]

As the next step towards becoming an industrialized nation, Vietnam will need to improve its rice cultivation technologies (technical facilities, fertilizers, seeds, irrigation system and farmers’ capacity).

However, up to now the handling of the Vietnamese Government lacks of activity in regard to mitigate the possible effects of As contamination of water and soils. In several areas the As concentration in drinking water is reduced in the waterworks due to iron elimination. But in rural areas no serious measures are taken to protect the rural population from the potential risk of As poisoning.

[8]Source: http://www.economist.com/news/asia/21594338-vietnams-farmers-are-growing-crop-no-longer-pays-its-way-against-grain

6 Conclusion

Our own investigations of soil, water, plants, wastewater and sewage sludge in the study area of Dai Lam and the review of more than 300 scientific references illustrates the multidimensional complexity of human exposure to As in this region of the Red River Delta. The origin of the As is geogenic, but there are many indications that the mobilization of As is a result of anthropogenic and hydrological circumstances. Bioavailability is a result of mobilization and of agricultural methods. The future development of As mobilization and distribution will be influenced by climate change and the socio-economically induced changes in water and land use.

To date, no comparable detailed investigation of As in the everyday activities of a distinct study area in the rural Red River Delta has been published. This might be a result of the relatively challenging circumstances when conducting such a study. The sampling campaigns and analyses required advanced technological equipment and experience, which were not self-available during the investigations. For example, various attempts were undertaken in the provincial capital of Bac Ninh to commission a laboratory with basic analysis, but in the end the water samples had to be analyzed by the Vietnam Academy of Science and Technology in Hanoi while all the other samples were transported to Germany, where they were prepared and analyzed at the Institute of Waste Management and Contaminated Site Treatment in Pirna. Accordingly, the number of samples was limited. It can be concluded from this that so far environmental monitoring of subjects of protection in rural areas in Vietnam has been doomed to failure because of the lack of analyzing know-how and technology.

In three water sampling campaigns in 2011, 2012 and 2013, it was shown that the shallow aquifer is subjected to As contamination. The average As concentration exceeds the international standard of the WHO of 10 µg/L. Moreover, the concentration in the wells is highly unsteady and so the potential risk for the individual households is difficult to assess. The household inventory showed that 170 out of 280 households (60%) still used the tube well water which originates in the shallow aquifer and 20% of these households didn't use any filters, which would substantially reduce As concentrations. This shows that although the village of Dai Lam is situated in the peripheral area of the highly contaminated region in the southeast of the Red River system, a significant proportion of the villagers are exposed to contaminated water.

The variation of As peaks in August, probably as a result of interfering climatic and human activities: the harvest and the following food processing activities lead to increasing wastewater quantities and to an increase in the organic matter in the wastewater. Until 2012, the wastewater was drained untreated into the irrigation system of the paddies, where high levels of organic matter led to anaerobic and thus reducing conditions. The As content in the paddies is not particularly high (6.34 – 33.0 mg/kg) and also SEF showed that those fractions that are supposed to bear the most bioavailable species (nonspecifically adsorbed and specifically adsorbed) only account for 10% of the As, the fractions which are associated with Fe and Al oxy hydroxyl minerals being the most abundant species. The particular circumstances in the paddy soils, which have been submerged for thousands of years, and which are probably accompanied by reducing processes most of the time, probably lead to the enhanced bioavailability of the fraction F3.

Organic matter enhances the growth and the metabolism of soil microorganisms which play a major role in the oxidation, reduction and methylation processes. Thus, the application of organic matter may increase the mobilization, volatilization and bioavailability of arsenic (Huang 2011). Moreover, soil properties such as pH and redox conditions may be affected by the change of organic matter and hence microbiotic activity (Zheng 2013). As an additional effect, changes to the organic matter level also affect the oxidation state of iron and subsequently the adsorption of As(V) on iron(hydr)oxides. These effects show that the application of fertilizers may have severe effects on arsenic release into the soil water and on the bioavailability of arsenic.

Precipitation and warm temperatures in July and August affect the horizontal and vertical mobilization of As. Due to the irregular dispersion of permeable layers and lenses, the concentration of As in the groundwater is scattered.

After the wastewater treatment facility of the INHAND project had been installed and put into operation, the As concentration in four wells revealed limited As concentrations and reduced variability, which could be a result of the decreasing release of organic matter into the soil.

The analyses of food samples and the review of international studies revealed that the daily diet bears potential risks of increased As uptake. Rice, meat and leaf vegetables are the main sources of As uptake and can be as high as 0.353 mg/d, which exceeds the PTWT of a person weighing 75 kg (0.15 mg/d) as well as the Codex Alimentarius threshold of 0.2 mg/d (As_{in}) and 0.3 mg/d (As_{tot}). Especially poorer people, whose diet relies on rice, leaf vegetables, offal and GAS, may be exposed to higher As concentrations.

The transition economy and its possible impact on human exposure to As

The study area is a typical food processing craft village in the Red River Delta. This system of local production, processing and consumption is widespread in Vietnam and bears a range of advantages: low transport and storage costs, and employment of the rural population. The craft villages are considered one of the pillars of the Vietnamese economy. Their development was probably an initial boost towards Vietnam's impressive economic growth, i.e. the craft villages are intrinsically linked to the transition economy. This explains why this system has been supported by the government. However, the rather uncontrolled development of the craft villages led to an unforeseen growth of pollution problems. No viable legal framework has been created yet to tackle the problems of water, soil and air pollution. The current directives comprise standards which are much too rigorous to be met, and for this reason measures for the protection of the protective goods are hardly implemented. An additional problem consists in the lack of responsibility. The infrastructure of the craft villages is assigned to the Ministry for Investment, the Ministry for Rural Development, the Ministry for Environment and

Natural Resources, the Ministry for Construction and the related agencies. The Tam Nong assigns supervision jurisdiction to various ministries or offices. However, so far no substantial improvement has been achieved.

The current situation for potential human As exposure turns out to be as follows (Figure 40). Paddy field cultivation leads to the increasing mobilization of As in soil. The insufficient wastewater treatment infrastructure and the drainage of wastewater with high organic content from food processing activities into the irrigation system produce increased reducing conditions and thus enhance the mobilization of As. Transport processes to the aquifer and As uptake in plants increase human exposure to As.

Seen from a medium to long term view, Vietnam and especially the regions adjacent to the economic and industrial centers Hanoi and Ho Chi Minh City will undergo strong changes in land and water use, which will probably affect the problems of As in the groundwater and crops.

Vietnam's economy is situated in the transition phase from a planned economy to a free market, from an agrarian nation to a powerful industrial state, and from a low level income to a middle level income country. The province of Bac Ninh, which belongs to the northern key economic zone, will be developed into an industrial region and thus become a driving force of the Vietnamese economy. The Vietnamese government has declared its intention to see many industrial zones built in this region and has also considered the growing numbers of inhabitants in the master plan. The plans imply substantial changes for water and land use such as the growth of sealed soil, built-up areas and forestry (Decision 198/QD-TTg). Moreover, Vietnam is among the countries which are badly affected by climate change in the form of land loss in coastal areas due to saltwater intrusion.

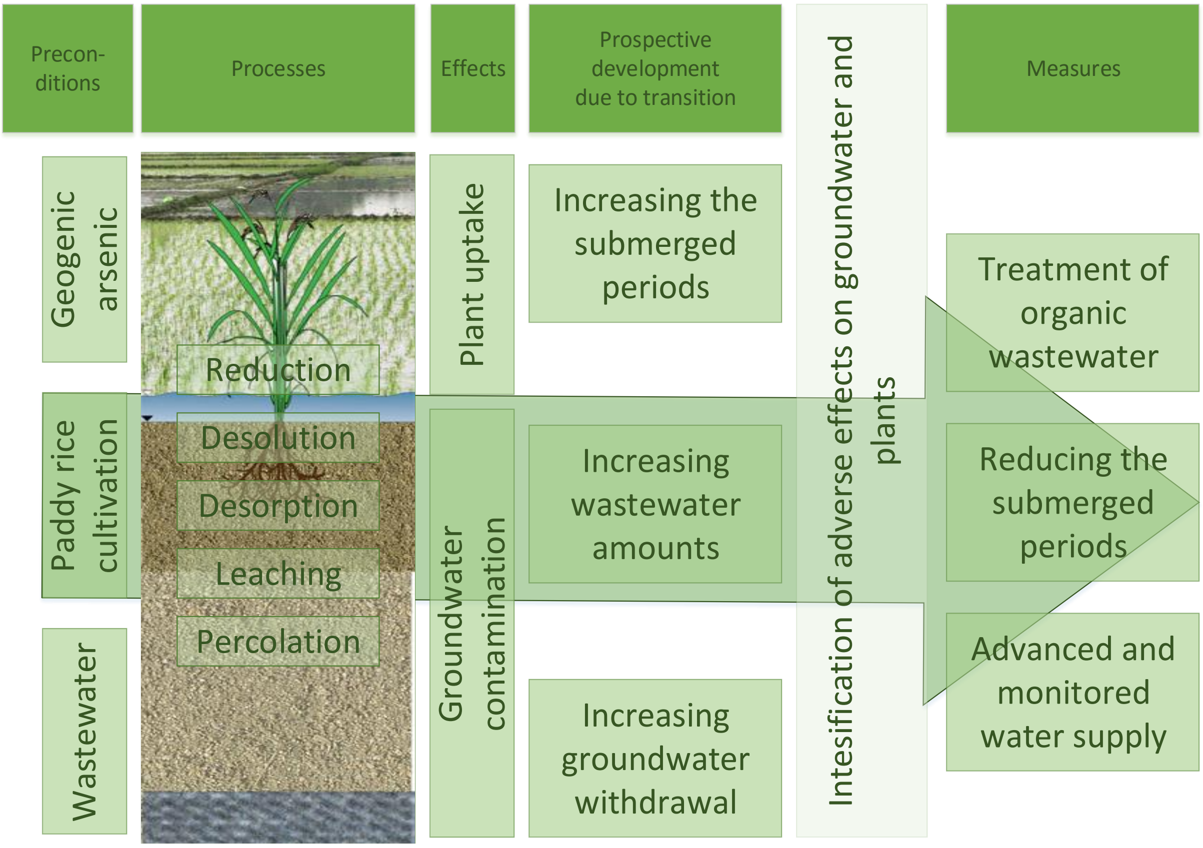

Figure 40: Key factors for human exposure to As in the Red River Delta, possible impact of the transition economy and mitigation measures

As a consequence, rice production will be intensified through greater mechanization, more pesticides and fertilizer, and increasing the number of harvests from two to three every year. An extra harvest will have several effects: (1) increased water consumption, (2) increasing greenhouse gas production (currently paddy fields produce 50% of Vietnam's methane (Rutten 2014), and (3) the mobilization of As which thus can enter the human food chain.

Moreover, industrialization and rising living standards will lead to increasing water withdrawal from the Pleistocene aquifer, with adverse effects on the As content in this currently unaffected aquifer (van Geen et al. 2013).

Although the higher water consumption isn't likely to lead to water shortages, the intensified use of groundwater and river water for irrigation will result in an unforeseeable intervention in the region's water balance.

Mitigation measures for human exposure to As in the Red River Delta due to transition and industrialization:

1. Reduction of organic wastewater drainage into the paddy fields by wastewater treatment facilities, especially in food processing craft villages.
2. Groundwater recharge of the Pleistocene aquifer.

3. Using cultivation methods like alternate wetting and drying (AWD), which have been proved to reduce the level of As in rice grains (Linquist et al. 2015).

7 Perspectives (further work)

Detailed investigations of the As behavior in paddy soils under consideration of changing environmental circumstances. Soil column experiments could deliver important results about the possible transport of As into the shallow aquifer.

Studies about the possible effect of anthropogenic activities, like increasing water withdrawal, increasing wastewater production and intensified agriculture.

Further structured investigation of as content in plants and vegetables, in order to create a reliable data pool for human risk in affected areas like the Red River Delta.

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9 Annex

9.1 Error Analysis

The results of an analysis campaign are subject to a certain impressision and must be reviewed critically. Each step is source of errors and uncertainty: sampling procedure, sample storage and transport, sampling treatment and the analyzing procedure. Every endeavor must be made to keep the errors to a minimum.

The errors are composed by a systematic and a random component. The random errors can always occur during a data acquisition and the cause can origin from the environmental conditions, from the measuring (reading) person or from the measuring instrument. The random error can be minimized by the accuracy and multiple measurements. The systematic error can occur by using measuring methods and measuring machines. Integrating calibrating measurements, recovery measurements are reducing the effect of systematic errors.

Measures to minimize the random and systematic analyzing errors:

The groundwater samples were taken by operating the electric pumps over 15 minutes. The samples were transported in cooling boxes. The samples were analyzed in as duplicates of approximately 10% of total samples.

The plant and meat samples were transported in cooling boxes to Pirna/Germany. Attempts were made to provide a reliable base for the samples: each plant sample consists of a mixture of several plants from each sampling point and the soil material as well as the animal products was mixed carefully before digesting.

Before each analyzing campaign the ICP-MS was recalibrated and each sample was analyzed in triple determination with threefold measurement. Possible interference occurs because of increased iron concentration in pure water.

9.2 Data

9.2.1 Pearson's correlation of As and five heavy metals in the upper soil samples

	Fe	Mn	Cr	Co	Cd
As	0.308	0.333	-0.044	0.127	0.419
	0.33	0.29	0.891	0.694	0.154
Fe		0.894	0.7	0.162	-0.106
		0.0000896	0.0112	0.615	0.744
Mn			0.349	0.136	0.141
			0.267	0.673	0.662
Cr				0.0709	-0.326
				0.827	0.301
Co					0.329
					0.296

9.2.2 Pearson's correlation of As and five heavy metals in the root zone samples

	Fe	Mn	Cr	Co	Cd
As	0.723	0.917	0.971	0.946	0.964
	0.008	2.7E-05	1.5E-07	3.35E-06	1.07E-07
Fe		0.813	0.777	0.778	0.625
		0.00129	0.00297	0.00288	0.0298
Mn			0.955	0.977	0.927
			1.4E-06	4.94E-08	0.000014
Cr				0.981	0.965
				1.97E-08	3.7E-07
Co					0.955
					1.29E-06

9.2.3 As in rice plants [mg/kg]

Samples	Leaves	Stems	Roots
1	7.67	2.19	51.39
2	8.22	6.12	63.37
3	5.02	1.70	28.69
4	7.67	1.95	65.39
5	8.22	5.75	65.87
6	5.02	1.65	29.55
7	5.23	2.03	68.18
8	7.41	5.84	67.95
9	4.64	1.65	26.58
Mean	6.57	3.21	51.89
SD	1.53	2.03	18.41

9.2.4 Heavy metals in wastewater samples

		Dec-11	Apr-12	Oct-12	Jan-13
Al	[µg/L]	3.9	3.5	6.2	2.5
Cr	[µg/L]	11.1	23.5	16.8	15.4
Fe	[µg/L]	5564.0	2514.0	2356.0	15247.0
Mn	[µg/L]	279.5	351.0	221.3	105.0
Co	[µg/L]	2.2	0.8	3.2	6.2
Ni	[µg/L]	29.0	26.0	42.8	25.4
Cu	[µg/L]	17.0	15.7	10.3	7.6
Zn	[µg/L]	67.3	102.0	80.2	78.7
As	[µg/L]	7.9	12.5	10.3	15.6
Cd	[µg/L]	0.1	0.1	0.1	0.0

9.2.5 Heavy metals in pig manure [mg/kg]

	manure 1 (industrial food)	manure 2 (rice fed)
Cr	4.1	263.8
Fe	1094	3160
Mn	419	441
Co	0.6	2.5
Ni	121.9	22.7
Cu	57.1	928.4
Zn	223	563
As	0.32	1.25
Cd	0.18	0.17
Pb	1.57	2.09

9.2.6 Heavy metals in pork liver and meat [mg/kg] ww

Liver	Pork 1	Pork 2	Pork 3	Pork meat
As	0.42	0.01	0.79	0.07
Cr	3.35	0.43	0.54	1.18
Fe	278.9	169.7	232.9	58.9
Mn	3.29	3.40	4.42	0.56
Co	0.03	0.04	0.04	0.01
Ni	0.24	0.18	0.73	0.21
Cu	48.8	23.6	97.4	2.71
Zn	182.3	142.9	204.3	92.6
Cd	0.05	0.04	0.23	0.02
Pb	0.03	0.12	0.49	0.04

9.2.1 Groundwater analyses

Table 27: Tube well analyses December 2011

Site number	T	pH	DO	EC	OPR	SO_4^{2-}	NH_4^+	NO_3^-	TOC	As	Fe	Mn	Pb	Coliform	Fecal Coliform
unit	°C		mg/kg	µS/cm	mV	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	MPN/100ml	MPN/100ml
1	24.7	6.8	4.01	1416	-111	29.63	7.92	0.26	11.2	0.01	6.61	0.16	0.00	2.00	ND
2	25.2	6.7	0.58	562	-126	<1	7.15	0.27	18.7	0.00	7.85	0.20	<0.001	ND	ND
3	25.6	7.0	1.11	1612	-198	99.30	3.45	0.14	8.4	0.02	4.26	0.09	<0.001	ND	ND
4	25.5	6.8	0.61	751	-139	1.20	5.85	0.19	15.6	0.00	7.98	0.17	0.00	ND	ND
5	25.7	6.9	1.32	859	-133	11.90	6.88	0.18	16.7	0.00	4.58	0.15	<0.001	ND	ND
6	25.0	6.6	1.15	672	-132	11.20	7.73	0.19	21.6	0.01	6.96	0.12	0.00	ND	ND
7	25.2	6.6	0.95	841	-145	<1	8.78	0.60	20.2	0.00	14.04	0.22	<0.001	ND	ND
8	25.4	6.8	1.77	811	-136	89.80	4.80	0.16	15.7	0.02	7.64	0.27	0.00	ND	ND
9	25.9	6.7	1.98	796	-156	<1	4.35	0.13	13.8	0.02	4.16	0.19	<0.001	15.00	ND
10	25.5	6.6	1.13	775	-133	<1	6.68	0.21	16.1	0.01	3.28	0.12	<0.001	4.00	ND
11	25.6	6.8	0.86	584	-188	<1	4.18	0.14	12.8	0.02	2.45	0.10	<0.001	ND	ND
12	25.5	6.6	0.84	634	-170	14.60	5.15	0.20	14.6	0.02	7.80	0.10	<0.001	3.00	ND
13	25.5	7.0	1.09	404	-128	<1	3.53	0.19	9.2	0.03	2.35	0.04	<0.001	4.00	ND
14	25.4	6.8	1.25	553	-126	<1	4.55	0.26	13.2	0.02	3.40	0.06	<0.001	3.00	ND
15	25.7	6.8	0.40	608	-155	<1	6.15	0.19	14.3	0.01	3.83	0.06	<0.001	2.00	ND
16	25.7	6.7	1.52	427	-139	<1	5.10	0.17	7.6	0.01	2.69	0.04	<0.001	ND	ND
17	25.7	7.0	0.55	417	-168	<1	3.75	0.22	8.0	0.01	1.04	0.02	<0.001	2.00	ND
18	25.7	6.8	0.81	675	-152	<1	5.30	0.23	11.5	0.00	3.44	0.11	<0.001	ND	ND
19	25.5	6.8	0.33	1152	-59	61.70	1.63	1.50	24.5	0.00	0.67	0.49	<0.001	3.00	ND
20	25.2	6.8	1.09	1156	-95	52.70	5.58	0.23	23.8	0.01	1.74	0.38	<0.001	2.00	ND

Table 28: Tube well analyses August 2012

Site number	T	pH	DO	EC	OPR	SO_4^{2-}	NH_4^+	NO_3^-	TOC	As	Fe	Mn	Pb	Coliform	Fecal Coliform
unit	°C		mg/kg	µS/cm	mV	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	MPN/100ml	MPN/100ml
1	26	7.45	6.55	1415	-115	38.7	7.54	0.16	3.78	0.02	33	1.08	0.001	7	3
1	27.6	6.75	2.01	1021	-69	42.3	6.13	0.71	16.32	0.008	0.425	1.11	0.001	4	2
2	25.9	7.07	1.19	587	-179	<1	6.53	0.12	6.47	0.006	39.8	1.27	0	3	ND
3	26	6.87	3.59	809	-162	79.9	5.85	0.15	11.52	0.069	25.9	0.693	0.001	4	2
4	26	6.7	2.08	1033	-185	1.13	5.13	0.14	5.12	0.01	67.3	1.61	0.003	4	1
6	25.8	6.89	2.78	750	-162.6	<1	6.58	0.16	8.67	0.007	40.4	1.49	0.001	9	3
7	26.1	7.22	3.19	882	-158.2	<1	7.75	0.14	11.52	0.015	45.1	0.988	0.002	2	1
8	26	7.06	0.29	933	-173.7	22.8	4	0.13	5.34	0.062	13.2	0.331	0.001	4	1
10	26	6.8	0.86	1196	-151.3	<1	3.98	0.37	17.87	0.047	15.5	0.389	0.001	3	1
13	25.9	6.98	0.76	750	-138.3	<1	5.3	0.09	7.5	0.071	18.5	0.517	0.064	7	4
14	26.7	7.2	2.25	481	-137.2	<1	4.08	0.17	3.12	0.074	40.3	0.576	<0,001	3	1
15	26.3	7.1	1.38	7.9	-159.8	<1	3.5	0.54	5.37	0.106	14.4	0.364	<0,001	4	1
17	25.5	7.24	0.33	820	-208.3	38.1	4.55	0.12	5.27	0.098	23.1	0.583	0.002	5	2
20	25.6	6.97	0.12	1094	-135.6	1.2	5.7	0.08	2.17	0.043	28.7	0.586	<0,001	4	1
21	25.9	7.39	0.08	465	-162.3	<1	3.98	0.37	17.87	0.047	15.5	0.389	0.001	3	1
22	25.7	7.13	0.22	557	-147.6	1.22	9.68	0.11	1.72	0.014	4.44	4.8	0.001	7	2
23	25.7	7.2	0.2	604	-136.1	<1	3.3	0.18	6.67	0.014	4.08	1.92	0.001	3	1
24	25.6	7.4	0.2	641	-137	<1	8.3	0.27	6.62	0.015	51.3	1.47	0	3	1
25	26	7.48	2.03	580	-200	<1	7.6	0.18	7.12	0.008	51.5	1.46	0.001	7	4
26	26	7.48	0.55	438	-159.2	<1	20.18	0.16	6.32	0.054	70.1	1.3	0.001	4	1
26	26	7.48	0.55	438	-159.2	<1	5.08	0.16	3.62	0.014	46.6	1.37	<0,001	9	3
27	25.2	7.3	0.21	1535	-90.2	44.05	7.28	0.11	3.81	0.08	29.3	0.8	<0,001	11	4
28	25.7	7.05	0.32	989	-127	48.7	5.25	0.13	3.12	0.071	22.8	0.925	<0,001	9	3
29	25.1	7	0.13	630	-98.5	31.3	5.25	0.18	10.44	0.099	33.7	1.59	<0,001	7	2
30	25.8	7.3	0.21	467	-180	<1	11.28	0.22	1.4	0.051	84.3	1.36	0.002	11	4

Table 29: Tube well analyses April 2013

Site number	T	pH	DO	EC	OPR	SO_4^{2-}	NH_4^+	NO_3^-	TOC	As	Fe	Mn	Pb	Coliform	Fecal Coliform
	°C		mg/kg	µS/cm	mV	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	MPN/100ml	MPN/100ml
1	23.3	7.32	3.63	597	-60	2	<0.01	0.67	3.78	0.001	18.6	0.419	<0.001	11	5
1	23.3	7.28	1.64	518	-47	<1.0	0.24	0.68	16.32	<0.001	0.774	0.347	0.009	1	0
2	25	7.52	1.3	530	-136	<1.0	5.98	0.2	6.47	0.001	22.8	0.603	<0.001	15	2
3	24	6.88	1.36	1061	-147	85	2.9	0.51	11.52	0.04	14.3	0.208	<0.001	7	0
4	22.2	6.47	1.6	950	-168	<1.0	7.98	0.43	6.02	0.007	40.2	0.593	0.034	11	2
6	23.4	6.74	2.33	760	-159.7	<1.0	7.65	0.53	10.68	0.007	29.6	0.589	<0.001	5	0
7	24.4	6.76	2.55	936	-163.4	<1.0	9.68	0.49	8.53	0.006	41.5	0.998	<0.001	11	2
10	24.3	6.85	2.46	648	-146	<1.0	7.43	0.49	7.64	0.008	21.7	0.501	<0.001	7	5
13	24.3	6.85	1.4	761	-150.5	<1.0	13.73	0.73	6.55	0.004	26.1	0.713	<0.001	13	5
14	24	7.02	1.06	451.4	-126	<1.0	5.6	0.32	4.57	0.024	17.1	0.357	<0.001	11	5
15	25.8	6.83	1.23	627	-166.4	<1.0	7.03	0.3	6.73	0.024	9.3	0.328	<0.001	15	2
21	24.5	7.1	1.46	435	-137.8	<1.0	4.23	0.51	8.64	0.052	6.46	0.102	<0.001	16	5
22	25.4	7.23	1.38	592	-106	<1.0	2.2	0.46	4.66	0.027	23.3	0.314	<0.001	5	0
23	24.1	6.68	2.11	650	-123.5	<1.0	3.6	0.41	7.13	0.032	11.2	0.15	<0.001	11	2
24	22.4	6.16	2.3	554	-113.7	1	2.5	0.53	3.86	0.002	3.32	0.115	<0.001	11	2
25	24.5	6.8	0.7	563	-148.6	1	2.38	0.53	10.54	0.045	15.2	0.228	<0.001	5	0
26	23.1	6.7	1.61	598	-98.5	<1.0	0.68	4.06	13.6	0.013	9.74	0.067	0.033	5	0
27	24	6.33	1	573	-163.6	93	8.45	0.76	1.42	0.002	1.87	1.93	<0.001	16	5
28	22.3	7.1	3.25	490	-80.4	13	1.15	1.79	4.85	0.001	11.1	0.302	0.084	14	2
30	22.4	6.86	1.06	371	-97.5	2	1.78	0.54	5.14	0.028	5.47	0.086	<0.001	11	0

In der Schriftenreihe „Beiträge zu Abfallwirtschaft/Altlasten“ des Institutes für Abfallwirtschaft und Altlasten sind folgende Bände erschienen:

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Band 96	Desintegration und anaerobe Verwertung bioabbaubarer Biokunststoffe	30,00
Band 97	10. Biogastagung Dresden - Anaerobe Biologische Abfallbehandlung 2015	30,00
Band 98	*n.n. (Veröffentlichung folgt)*	
Band 99	Entwicklung und Implementierung einer Methodik zur Erfassung der Grünschnittpotenziale von Siedlungs- und Verkehrsflächen in kommunale Verwertungsstrukturen	30,00
Band 100	Review of arsenic contamination and human exposure through water and food in rural areas in Vietnam Hanoi	*30,00*

Die vergriffenen Bände 16, 27, 31,32 und 39 können als CD zum Preis von 15,- € + Porto und Verpackung versendet werden.

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